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Adsorption of methylene blue from an aqueous dyeing solution by use of santa barbara amorphous-15 nanostructure: Kinetic and isotherm studies

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Abstract. Santa Barbara Amorphous-15(SBA-15) nanoparticles were utilized as the inexpensive and effective adsorbents to remove methylene blue dye from the aqueous solution.SBA-15 was created by Zhao et al method. Infrared spectroscopy, X-ray diffraction and scanning electron microscopy (SEM) were used for the evaluated physical properties of SBA-15. The results of diffraction X-ray indicated that was the crystalline structure for it. Also IR spectroscopy indicated was a silica the whole structure of the groups and SEM image verify the structure of relatively identical particles size of SBA-15. Factors affecting adsorption including the amounts of adsorbent, pH and contact time were investigated by a SBA-15 nanomaterial design. The extent of dye removal enhanced with increasing initial dye concentration and pH from 4 to 10. The higher percentage adsorption were obtained under optimum conditions of variables (sorbent dose of 200 mg/liter،initial MB concentration 10 mg/liter, initial pH of 10 and temperature of 25°C). Maximum adsorption happened after the 2 hour and the kinetic processes of the dyes adsorption were described by applying the pseudo-first-order and the pseudo-second-order and the relatively High correlation with the kinetic Ellovich models. It was found that the pseudo-second-order models kinetic equation described the data of dye adsorption with a good correlation (R2>0.999) which indicated chemisorption mechanism. Freundlich and Langmuir adsorption models were investigated in conditions of variables (adsorbent dose 0.01 gr/liter, MB concentration 10, 20, 30 mg/liter, pH of 4, 7, 10, contact time 90 min and temperature of 27°C). The adsorption data were represented by Langmuir isotherm model. These values are higher than the adsorption capacities of some other adsorbents that have recently been published in the literature.

Keywords: isotherms adsorption; nanomaterial; SBA-15; cationic color; removal color

1. Introduction

Industries such as textile, cosmetics, and paper and pulp, are known for producing large volumes of colored effluent, which are difficult to treat due to the high chemical stability of dyes.

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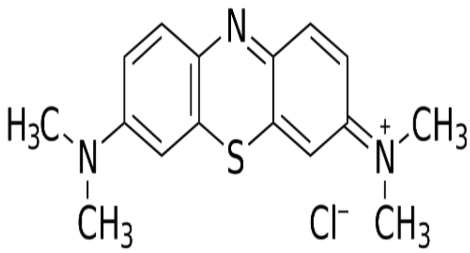

Fig. 1 Chemical structure of methylene blue

Table 1 Studies of SBA-15 in remove of color

Source	Adsorbent
Badiei *et al.* (2014)	Silica nanoparticles with amino groups
Huang *et al.* (2011)	SBA-15 silica Mesopore
Paul and Bhaumik (2012)	MBS5[†] and SBA-15
Chen *et al.* (2012)	SBA-15 with carboxylic functional groups

Table 2 Mesoporous silica material (Zhao *et al*. 2012)

Size of holes	Space group	Pattern	Precursors	Mesoporous silica materials
2-10	P6mm	CTAB, C_n TMA^+ (n=12-18)	TEOS, sodium silicate	MCM-41
5-30	Im3m	F127, F108, or F98	TEOS, TMOS	SBA-16
10-50	Cellular foam	F127 with TMB	TEOS	MCF
2-4	Ia3d	CTAB, C_n TMA^+ (n=14-18) $C_{16}H_{33}(CH_2C_6H_5)$	TEOS, sodium silicate	MCM-48
5-30	P6mm	B50-1500 ($Bza0_{10}$ EO_{16}), P123, P85, P65, Brij97($C_{18}H_{35}EO_{10}$)	TEOS, sodium silicate	SBA-15
2-10	Wormlike	$C_mH_{2\ m+1}$ NH_2 (m=8-22)	TEOS	HMS
4-27	Fm3m	F_{127} ($EO_{106}PO_{70}EO_{106}$)	TEOS	FDU-12
2-15	Fm3m	CTAB, OTAB, CPB, P123, F127, Brij56, Brij76	$(RO)_3$ Si-R-Si $(RO)_3$	PMO_S

Conventional physicochemical and biological treatment processes often prove inadequate to meet the discharge standards of different countries. Some of the treatment processes like membrane filtration, electrochemical, photo oxidation, and chemicals such as Fenton reagent or hypochlorite have been used to treat wastewater containing dye. However, all the processes have their own demerits associated with cost and sludge disposal dyeing industries effluents has many poisonous effects on aquatic ecosystems (Mane *et al*., Mall *et al*. and Srivastava *et al*. 2007). The colors and products resulting from their nondegradable and have toxic effects and carcinogen that creates numerous ecological problems. Aromatic rings in the structure of azo colors cause toxicity of these colors and it makes from the aspect of biological non-biodegradable (Xiao *et al.* 2015). As a result, this material cause be carcinogen and jumps to humans and aquatic organism (Hou *et al.* 2012). As a methylene blue azo cationic dye, with molecular formula C16H18N3SCl and molar mass of

[†]Borosilicate Material

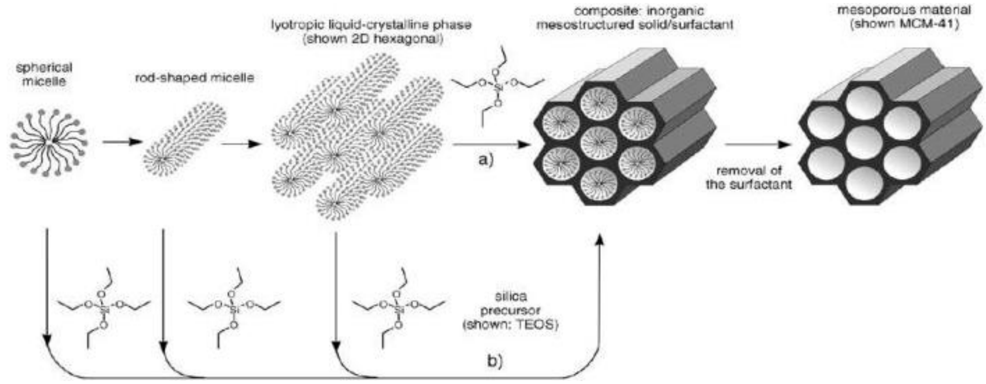

Fig. 2 The formation of mesoporous silica materials (Zhao *et al*. 2012)

319.85 g/mol is widely used in the industries of cotton dyeing, wool, silk, leather, hair color. The color can causing harmful effects such as irritation eye, hemoglobinemia Matt, convulsions, cyanosis, palpitations, shortness of breath and skin irritation in humans. Thus color removal from Industrial hysteresis like the methylene blue is the one of major ecological challenges. Methylene blue structure is shown in Fig. 1.

Adsorption process for non-biodegradable colors is very important. Nano structure adsorbents with a high specific surface and high adsorption power produce less lesions. The use of adsorption process in the high performance, possibility of recovery and re-use adsorbent, is economic justified. The silica mesoporous materials (MCM and SBA) of molecular sieves because of the surface area, high porosity, pore size distribution and homogeneous is considering control able level. This material has to member of the family properties good option for use as adsorbent, catalysts and so forth. Such applications can be changed to enhance the inside walls of the channels or connect the various a factor groups onto the surface of the material has founded through synthesis (Asouhidou *et al.* 2009), some recent studies in the field of color removal by effects of SBA-15 and Mesoporous silica materials were in Tables 1 and 2.

SBA-15 is a noticeable for remove of a cationic color. The purpose of this study nanoparticles SBA-15 synthesize for removal of the cationic methylene blue dye of aqueous solution. Chemical characteristics, structure, morphology and crystal structure SBA-15 by using the Infrared spectroscopy scanning electronic microscopy and X-ray diffraction pattern were identified. Adsorption of methylene blue on SBA-15 in various environmental conditions in terms of contact time, adsorbent dosage, pH and initial concentration of dye soluble was evaluated.

2. Materials and method

2.1 Chemical material and instrument

All chemicals material used were of analytical grade and demineralized water was used for the preparation of all aqueous solutions. Methylene blue powder(MB), PluronicP123,tetra-ethyl orthosilicate, chloric acid, sodium hydroxide was supplied by Merck(Germany). Also for the preparation and analysis by samples by of the devices magnetic stirrer (IKA RH basic2), a spectrophotometer (Biochrum Libra S22) for reading the concentrations, X-ray apparatus (Philips-

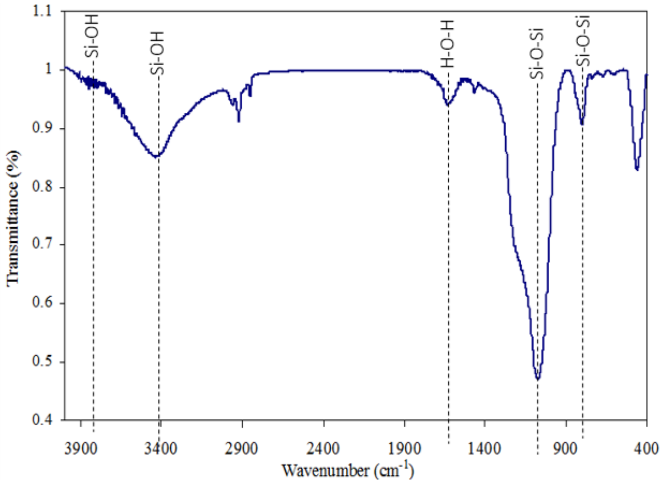

Fig. 3 IR spectrum of SBA-15

PW 17C diffract meter), transitional IR spectrometer instrument (Shimadzu 4600 spectrometer), scanning electron microscopy (Cambridge S-360) and centrifuges (Labnet 24D & SH12) reactions for the synthesis of adsorbing and separating its after adsorption were applied.

2.2 Making adsorbent and solution

In this research was used of Zhao substance for the manufacture of SBA-15 (Zhao *et al.* 1998). In this way, of the three-block copolymer Pluronic P123 was used (Aldrich; EO20- PO70- EO20). In this way was solved 4 gr of the Pluronic P123 in 30 gr deionized water and 120 g of H Cl (2 mole/liter) solution at the 35°C of temperature, then 5.8 gr of tetraethyl orthosilicate was added to the solution for 20 h at 30°C and stirred. Then the mixture was placed for about 24 hours at 80°C under quiescent conditions. The resulting solid was filtered and washed several times to overnight and was placed at room temperature to perfectly dry. Pluronic mesoporous shaping isolated with calcination was used at 6 hours and heating rate of 550°C/2 min. characteristic of SBA-15 samples washed with 30 ml of water. The Particle diameter size for SBA-15 manufacture in range of nanometers has been achieved. Dye solution manufactured by initial concentration of 240 mg/l in 27°C. Colour laboratories samples manufacture in concentrate of 10, 20, 30 mg/l. Formation of the mesoporous silica materials was shown in Fig. 2.

2.3 Theory of adsorption

The initial soluble with a concentration of 200 ppm of MB dye by dilution concentrations of 10, 20 and 30 mg it was prepared. All experiments by adding of the 50 ml dye soluble with 240 min of contact time and centrifuged for 5 min at the 40 rpm according to was conducted the method at time factor one. Specified optimal conditions to simultaneously enter and was determined optimum values for each parameter. By using of the spectrophotometer method λ max methylene blue 667 nm was obtained. That was studied different condition and effecting factors in the adsorption process. The initial concentration (10, 20 and 30 mg/liter), at various pH from 4 to 13 and the amount of adsorbent (40-400 mg/liter) was used. At the end of every experiment

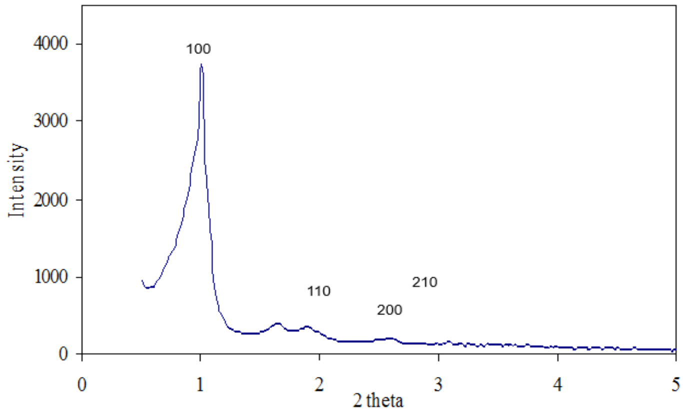

Fig. 4 X-ray diffraction pattern of SBA-15

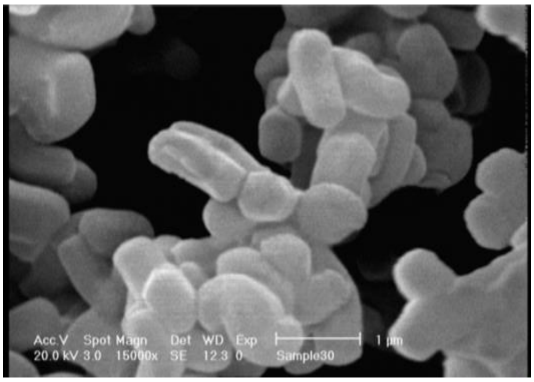

Fig. 5 SEM image calcined of hexagonal SBA-15 (the magnification×15000)

percentage removal of dye and the amount adsorbed (mg/g) determined by the mass balance equations.

3. Results and discussion

3.1 CharacterizationSBA-15

For investigate the functional groups attached to the SBA-15 adsorbent surface and study of the surface morphology used the FT-IR, XRD and SEM. (Figs. 3, 4, 5). Nanoparticles can be observed in the range of adsorption spectrum of 780 cm^{-1} (Badiei *et al.* 2014). In wavelength range of 1070-1220 related to symmetric and asymmetric tensile adsorption observed Si-O-Si group and siloxane clause. In IR spectrum of SBA-15 observed a wide band of 3745 cm^{-1} Si-OH group on the silica surface (Benhamou *et al*. 2009), hydroxyl group in 3400 cm^{-1} (Shahbazi *et al*., Younesi *et al*. and Badie *et al*. 2011) and in 1640 cm^{-1} the spectrum hydroxyl group (H-O-H).

For a better view peak of SBA-15 in the range of 0 to 5 degrees X-ray diffraction image of SBA-15 observed in Fig. 3. The peak of relevant for the crystal plates of SBA-15 sample observed in density of (100), (110), (200) and (210) that was of 2θ=0.6-2.07 (Zhao *et al.* 1998) which

Table 3 Physical properties of SBA-15

Sample name	Textural properties			Structural properties	
	S_{BET} (m^2/g)	V_P (ml/g)	d_p (A°)	d_{100} (A°)	b_p (A°)
SBA-15	479	0.51	35.8	87	32.4

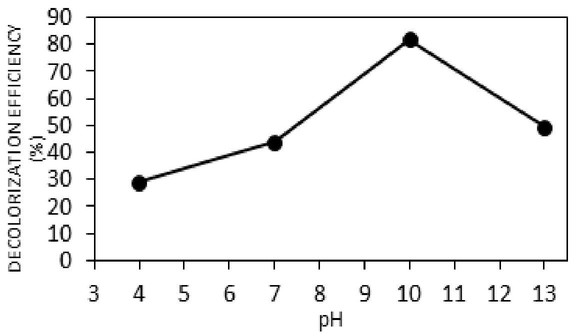

Fig. 6 The effect of pH in the removal rate of methylene blue

represents the regular hexahedron structure of SBA-15 is the formation of 100 represents the peak of symmetrical cavity at structure (Yue *et al.* 1999).

SBA-15 silica Nano pore SEM image observed at a magnification of 1 micrometer at Fig. 4. Surface morphology of SBA-15 was formed with a large number of clustered shape relatively uniform size and shape (average size of approximately 8.0 micrometers) and a lot of string gatherings (Chowdhury *et al.* 2011). The shape and internal morphology of SBA-15 particles sample was the hexahedron (Kresge *et al.* 2008).

The specific surface area, pore volume and pore size of the samples shown in Table 3. The BET surface area of the SBA-15 sample was 479 m^2 g^{-1}. This may be due to the polymerization of monomers inside the channels of SBA-15.

3.2 -pH

Quantity of pH is an important factor such as surface charge absorbent in solution ionization functional groups (Han *et al.* 2006). In this research study of pH on the absorption process in the range of 4 to 10 (4, 7, 10) and in terms of initial concentration of 20 mg/liter, adsorbent dosage of 200 mg/liter and 240 minutes contact time was evaluated. To adjust the pH, 0.1 Normal Hydrochloric acid and profits were used. The results show that further removal of methylene blue by SBA-15 at pH of 10 (Fig. 6). With increasing pH, amount hydroxyl ions in solution is made up which makes to adsorbent surface, negatively charged protons lost the and absorbing surface, and amount of power used to reinforce the electrostatic adsorption between color that are having a positive charge, and the surface increased absorbent followed by will attract more colors (Crini *et al.* and Badot *et al.* 2008). Given that the silica Nano pore because the presence of groups Si-OH, a negatively charged the surface, which prevents the attract negatively charged ions and cationic ions to attract more applications. So nanoporous silica performances in the processes of absorption are heavily dependent on functional groups on the surface nano-silica network to attract particular type pollutants.

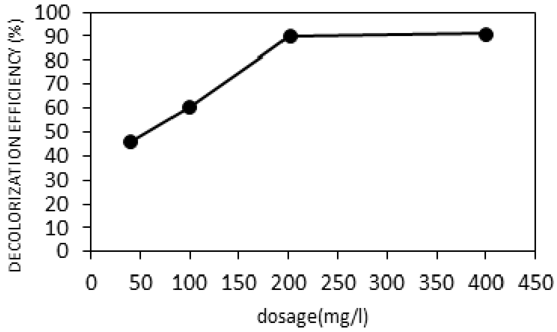

Fig. 7 Amount of adsorbent on the removal of methylene blue

Table 4 Effect of the adsorbent dosage on the adsorption process of methylene blue on the surface SBA-15

Adsorbent dosage mg/l	Secondary concentrations mg/l	Amount of attracted mg/g	Descoloration yield %
40	10.8	229.78	45.95
100	7.9	120.91	60.45
200	1.98	90.06	90.06
400	1.7	45.74	91.49

3.3 Adsorbent

To investigate the effect of adsorbent dosage was employed, different values of 40, 100, 200 and 400 mg/l of adsorbent in terms of the initial concentration of 20 ppm, pH of 10 and contact time 240 minutes. Increase the level of adsorbent and adsorption amount greater access to locations (Asadi *et al.* 2012) because an increased level of active catalyst, the amount of is also increasing decolorization (Ghanbarian *et al.* 2008). The reason for this theme be expressed, Overlap locations on the adsorbent surface is absorbed and thus reduce the yield leading to reduced absorbed and absorption rate locations has been (Mirhasani *et al.* 2014) with increasing the amount of absorbent than 200 to 400 mg/l of adsorption amount is reduced because increasing the amount of adsorbent to adsorb increases the number of free places, As far as the whole color molecules adsorbed on the surface of activated sites more the adsorbent and adsorbent increase (constant dye concentration) resulting in further increase free surfaces, causing it gets remain empty. That was percentage adsorption, remained unchanged equilibrium sorption capacity (Qe) is reduced (Table 4) (El Haddad *et al.* 2012).

3.4 Color change

There is a high correlation between the amount of the color initial concentration and remove it. The effects of amount initial dye concentration of color removal with the number of places on the surface the adsorbent to remove the color is associated. In the general case by increasing the color concentrations, rate of the color removal is reduced because of the charging of sites available for higher concentrations. By increasing color saturation of 10, 20 and 30 mg/liter in the conditions, pH=10 and adsorbent dosage of 200 mg/l and removal rate contact time of 150 minutes respectively 100, 87 and 74 percent. The number of adsorption sites on the surface of the

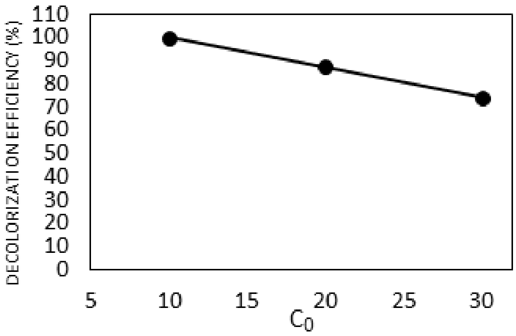

Fig. 8 Reduse color by initial concentration change

Table 5 The uptake kinetic model of Methylene blue with different adsorbent SBA-15

R^2	Kinetic Model	Adsorbent
0.999	Pseudo-second	SBA-15
0.993	Pseudo-second	SBA@Tris(2-aminoethyl)amine
0.999	Pseudo-second	SBA/HPG-COOH

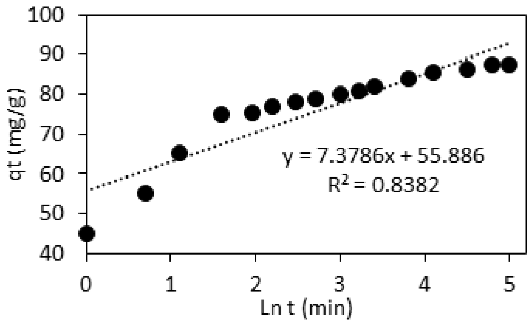

Fig. 9 Ellovich adsorption models of methylene blue on the SBA-15

adsorbent soluble is constant. Thus, with the increasing color concentrations of, the number of places available is less occupied and therefore has declined efficiency of the process (Li *et al.* 2013).

3.5 Kinetic

Kinetics of this reaction were examined with a 20 ppm of methylene blue, pH=10, 200 mg per liter adsorbent, in ambient temperature of 27 Celsius and sections according 1, 2, 3, 5, 7 , 9, 12, 15, 20, 25, 30, 45, 60, 90, 120, 150 minutes. The results of indicate that rate of adsorption for methylene blue on SBA-15 is very high. Such a way was seen 75% adsorption of methylene blue in 5 minute. The kinetic result was studied by three pseudo first-order, pseudo second-order and Ellovich. The results of indicate the correlation uptake kinetics with pseudo-second-order models. Previous studies of adsorption methylene blue kinetics is in Table 5.

3.5.1 Ellovich model

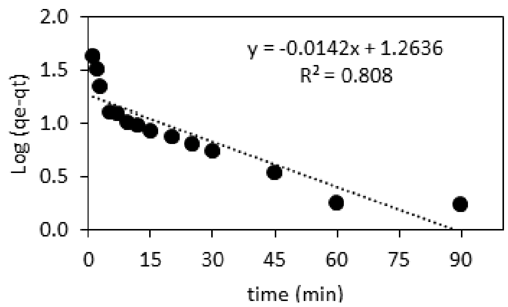

Fig. 10 Pseudo First-order adsorption models of methylene blue on the SBA-15

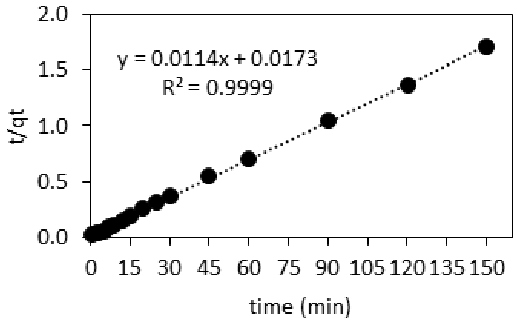

Fig. 11 Pseudo second-order adsorption models of methylene blue on the SBA-15

Study uptake kinetics results are represents relatively lower correlation with the adsorption rate methylene blue on the ellovich model SBA-15 (Fig. 9). Pseudo first order kinetics model Results of this study indicate a relatively low correlation with the pseudo first order kinetics of studies (Fig. 10). This model to describe the processes of adsorption in the solid-liquid phase has been relying on solid capacity (Li *et al.* 2015).

3.5.2 Pseudo second order kinetic model

Adsorption represents perfect correlation with the adsorption reaction is Pseudo-second kinetics model (Fig. 11). In this model, the rate of adsorption of the soluble pollutants at the beginning of the reaction was high as time went by the number of locations occupied in adsorbent surface increases, and the number of empty places for placement of pollutants-has been reduced and rate of absorption is reduced.

4. Adsorption isotherm

Equilibrium relationship between absorbent and adsorbates can be described by the adsorption isotherm. The relation between amount of material attracted and the remaining amount is usually in solution at the point of equilibrium and be expressed at a fixed temperature (Khaled *et al.* 2009). The research for the study of the adsorption isotherm, Freundlich and Langmuir models was used. Based on studies conducted in conditions of (maximum adsorption value 86.95 mg/g adsorbent quantity 0.01 g/liter, initial concentration of 10, 20 and 30 mg/liter, pH of 4, 7, 10 and contact time 90 min) was obtained.

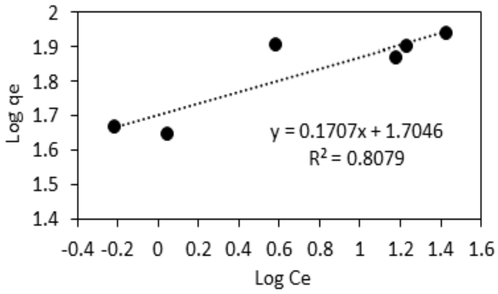

Fig. 12 Freundlich adsorption models of methylene blue on the SBA-15

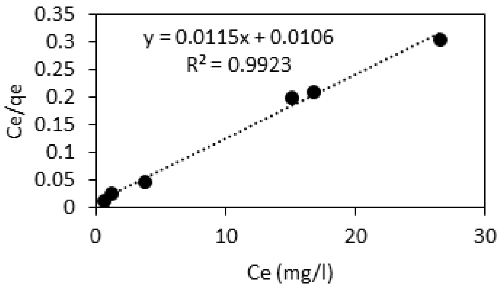

Fig. 13 Langmuir adsorption models of methylene blue on the SBA-15

Table 6 The adsorption isotherm model of methylene blue with different adsorbent SBA-15

R^2	Isotherm model	Adsorbent
0.987	Freundlich	SBA-15
0.999	Langmuir	SBA/HPG-COOH
0.998	Langmuir	GO-SBA-15

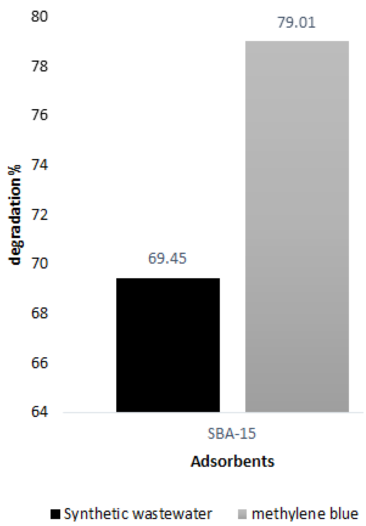

Fig. 14 Removing of the methylene blue comparison in optimum conditions (adsorbent of 200 mg/l, pH of 10, initial dye concentration of 20 mg/l and contact time 120 min) by MB pure and solution of MB in pH=10 and many component include many heavy metal and $Na_2S_2O_4$

4.1 Freundlich isotherm

The results are match with Freundlich isotherm models shown fairly low adsorption (Fig. 12). Freundlich model is an empirical model that could to be used to describe the heterogeneous adsorption systems.

4.2 Langmuir isotherm

The results of the tests with the Langmuir model represents a significant correlation (Fig. 13).

Langmuir isotherm for adsorption of dissolved substances in a single layer on adsorbent surface with number of available places is a limited. With regard to single-layer structure of SBA-15 and adsorption mechanisms by available places on it, the Langmuir model's ability for describe of the methylene blue adsorption is very high. In the Table 6 followed similar studies of adsorption isotherm model for methylene blue with different adsorbent SBA-15 family.

5. Conclusions

In results ability of the SBA-15 nanostructure for adsorbed of methylene blue dye was shown. The most removed with 200 mg/liter adsorbent dosage, 10 mg/l of the initial concentration, pH of 10 and contact time 120 minutes to much as 98 percent and also was the contact time of 150 minutes to 100%. This results shown that the uptake kinetic of pseudo second-order model and temperature model also complies with Langmuir model. In this study the performance of nano adsorbents for the removal of sample color of methylene blue and optimum condition of nano adsorbents for removal of the effluent was studied.

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