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Removal of cephalexin from artificial wastewater by mesoporous silica materials using Box-Behnken response surface methodology

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ABSTRACT

Mesoporous silica such as MCM-41 was used as an adsorbent for the removal of cephalexin antibiotic from synthetic wastewater. In this study, MCM-41 adsorbent was synthesised and was characterised by FE-SEM, XRD, FTIR and isotherms of adsorption/desorption of N_2. The effects of initial pH, adsorbent dose, initial adsorbate concentration, contact time and temperature on process efficiency were evaluated using Box-Behnken statistical experiment design (RSM). FTIR analysis revealed the Si-OH, H-O-H and Si-O-Si bonds are formed. According to BET surface area, MCM-41 had pores with a diameter of more than 2.0 nm and surface area of 1,097 m^2g^{-1} and also XRD spectra showed the mean crystallite size of MCM-41 was 75 nm. The statistical results show that pH, adsorbent dose, initial antibiotic concentration, temperature and quadratic pH were significant and presented with probability <0.05. The optimum removal condition based on analysis of variance and the quadratic model was the initial pH of adsorbate solution fixed at 3.00, adsorbent dose 800 mg L^{-1}, the initial concentration of antibiotic at 50.0 mg L^{-1}, a temperature of 40.0°C, and at the adsorption time of 30.0 min. Under these conditions, the percentage removal of cephalexin antibiotic was 90.3%. Therefore, according to the obtained results, the mesoporous silica can be used to adsorb cephalexin antibiotic in optimal conditions designed by response surface methodology.

Keywords: Adsorption; Cephalexin; Mesoporous silica; Box-Behnken response surface methodology

1. Introduction

With the development of different industries, a large volume of emerging contaminants is generated. These contaminants present unknown effects, short, medium or long-term effects on the environment and human health [1]. The effects of the residues of emerging contaminants left in the environment are not entirely understood, and they do not present values of maximum permissible concentrations in the environment yet [2]. The emerging contaminants includes chemical compounds used in the preparation of cosmetics (creams, perfumes), surfactants, degreaser, pharmaceuticals, pesticides, plasticisers, flame retardants and other compounds [3,4].

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Antibiotics are a group of pharmaceuticals that extensively used in medicine, animal husbandry and agriculture [5]. These pharmaceuticals could participate in the water cycle in two primary ways [6]. Firstly, the pharmaceutical industry, which includes four different types of manufacturing processes, fermentation, chemical synthesis, extraction and formulating [6]. The second way is the human releases from their excretions into domestic sewage considering that the biological treatment of sewage is not complete; the pharmaceuticals can be released into wastewater in metabolised or non-metabolised form. Most of the antibiotics are not biodegradable [7–10].

Recently high concentrations of antibiotics were found in several environmental compartments that expose risk to human lives [11–13]. They commonly could increase the tolerance of bacteria's causing risk to humanity. Cephalosporins are antibiotics with the same basic structural requirements as penicillin [10–12]. These antibiotics are utilised to therapy of a variety of infections in human [14]. Cephalexin has widely used for therapy of different infections in human due to gram-positive and gram-negative bacteria [14–16].

Since antibiotics are overused, recently they were observed in flowing out of hospital wastewaters, effluent and wastewater treatments plants at mg L^{-1} levels [17]. Therefore, it is essential to develop new methods to effectively remove these contaminants from aqueous solutions using technologically available and economic conditions [12,18–20].

These methods include chemical oxidation [21], biodegradation [22] and physical processes [23,24]. Adsorption is the most applied physical-chemical processes of removal of emerging contaminants from aqueous solutions [25–29], due to its low initial cost for implementation, simplicity and flexibility of the system, easy operation [22,30–33].

Various adsorbents have been recommended for the removal of pharmaceuticals such as activated carbon [18,19], zeolites [34], clays [35] and mesoporous silica [36,37]. Among mesoporous silica, the MCM-41 silica is a novel mesoporous that presents a large surface area and nanometer-sized pores (from 2.00 to 5.00 nm), high surface area (up to 1200 m^2 g^{-1}), and large and identical pore size, tunable pore structure and environmental compatibility [38].

Response surface method (RSM) is a mathematical and statistical technique which is useful for the optimisation of chemical reactions and industrial processes and is also commonly used for experimental designs. In statistics, response surface methodology explores the relationships between several explanatory variables and one or more response variables. The main idea of RSM is the use of a sequence of designed experiments to obtain an optimal response [39–45].

The aim of this study was the removal of cephalexin antibiotic from aqueous solution by MCM-41 mesoporous silica adsorbent using the statistical response surface methodology (RSM) to optimisation of adsorption conditions, modelling of the adsorption process and saving of time.

2. Materials and methods

2.1. Solutions

In this study, cephalexin antibiotic (with a purity of 99.8%) was acquired from Sigma-Aldrich and used without further purification. A stock solution of cephalexin (1.00 g L^{-1}) was prepared with deionised water and diluted as necessary. The chemical structure of the cephalexin is available in Fig. S1.

2.2. Preparation of MCM-41 silica

Mesoporous silica was prepared by following protocol [46]. An amount of 1.00 g of $C_{16}H_{33}N(CH_3)_3Br$ (CTAB, cetyltrimethylammonium bromide, Acros) was dissolved in 500 mL of water, and then it was added 3.50 mL of 2.00 mol L^{-1} of NaOH (pH value was about 12.0). The mixture was heated to 80.0°C, and then it was added to 5.00 mL of tetraethyl orthosilicate (TEOS, $Si(OC_2H_5)_4$, Acros) slowly. Subsequently, the TEOS was added, the system was magnetically stirred for 2 h at 80.0°C. Further, it occurred as a formation of a white solid that was separated by centrifugation at 12,000 rpm. The solid was thoroughly washed with water, followed by methanol, to eliminate the surfactants from the product. Finally, the solid was dried under reduced pressure (50.8 mm Hg at 70.0°C) for atleast 15.0 h. Afterwards, the adsorbent material was calcinated at 550°C for 5.00 h to remove the residual of CTAB used during the synthesis of MCM-41 mesoporous silica.

2.3. Characterisation of MCM-41

In this study, the specific surface area and pore size distribution were determined by nitrogen adsorption/desorption analysis. It was employed X-ray diffraction analysis to confirm the mesoporous silica crystalline structure. FT-IR analysis was used to verify the releases of the surfactant before and after calcination.

2.4. Experimental design

A Box-Behnken response surface methodology (RSM) and Minitab software were used for experimental design and evaluate the effects of five independent factors that could influence the batch adsorption system, respectively. These factors were pH (A), the dosage of adsorbent (B), the initial concentration of antibiotic (C), time of adsorption (D) and temperature (E). The utilisation of RSM supply a mathematical relationship between factors and the experimental results that can be fitted to a second-order polynomial model as Eq. (1).

$$\begin{aligned}\text{Response} = {} & \beta_0 + \beta_1 A + \beta_2 B + \beta_3 C + \beta_4 D + \beta_5 E + \beta_6 AB \\ & + \beta_7 AC + \beta_8 AD + \beta_9 AE + \beta_{10} BC + \beta_{11} BD \\ & + \beta_{12} BE + \beta_{13} CD + \beta_{14} CE + \beta_{15} DE + \beta_{16} A^2 \\ & + \beta_{17} B^2 + \beta_{18} C^2 + \beta_{19} D^2 + \beta_{20} E^2\end{aligned} \quad (1)$$

In this equation $\beta_1, \beta_2, \beta_3, \ldots \beta_{20}$ are coefficients of the regression equation and A, B, C, D and E, are the factors described as follows.

The levels of the factors based on the experimental design are presented in Table 1. In this model, the experiments were utterly random, and the total of experiments for designing and reviewing five factors in three levels and six replications at the central point was 46 independent experiments. The R^2 and R^2_{Adjusted}, the normal distribution of the residuals and the plot of actual values vs. predicted values were obtained to determine the quality of the proposed model. Analysis of variance (ANOVA) was used as a method of statistical analysis of responses, and the corresponding plots were designed

Table 1
Variables, levels of design experiments and Box-Behnken design (5 factors, 46 experiments performed)

	Factor	Low level	Central point	High level
Coded		−1	0	1
pH	A	3.00	7.00	11.0
Dose MCM-41 (mg L^{-1})	B	200	500	800
C_0 cephalexin (mg L^{-1})	C	50.0	75.0	100
Time (min)	D	30.0	60.0	90.0
Temperature (°C)	E	20.0	30.0	40.0

Exp	A	B	C	D	E	% Removal	Exp	A	B	C	D	E	% Removal
1	3.00	200	75.0	60.0	30.0	54.1	24	7.00	800	100	60.0	30.0	45.2
2	11.0	200	75.0	60.0	30.0	8.92	25	3.00	500	75.0	30.0	30.0	60.6
3	3.00	800	75.0	60.0	30.0	72.2	26	11.0	500	75.0	30.0	30.0	12.2
4	11.0	800	75.0	60.0	30.0	17.2	27	3.00	500	75.0	90.0	30.0	68.2
5	7.00	500	50.0	30.0	30.0	39.2	28	11.0	500	75.0	90.0	30.0	14.9
6	7.00	500	100	30.0	30.0	24.7	29	7.00	500	50	60.0	20.0	39.9
7	7.00	500	50.0	90.0	30.0	44.4	30	7.00	500	100	60.0	20.0	25.5
8	7.00	500	100	90.0	30.0	29.3	31	7.00	500	50.0	60.0	40.0	43.8
9	7.00	200	75.0	60.0	20.0	21.2	32	7.00	500	100	60.0	40.0	26.8
10	7.00	800	75.0	60.0	20.0	47.4	33	3.00	500	75.0	60.0	20.0	63.2
11	7.00	200	75.0	60.0	40.0	22.3	34	11.0	500	75.0	60.0	20.0	12.8
12	7.00	800	75.0	60.0	40.0	48.9	35	3.00	500	75.0	60.0	40.0	66.8
13	3.00	500	50.0	60.0	30.0	69.7	36	11.0	500	75.0	60.0	40.0	14.3
14	11.0	500	50.0	60.0	30.0	15.9	37	7.00	200	75.0	30.0	30.0	20.5
15	3.00	500	100	60.0	30.0	56.5	38	7.00	800	75.0	30.0	30.0	45.9
16	11.0	500	100	60.0	30.0	11.0	39	7.00	200	75.0	90.0	30.0	23.0
17	7.00	500	75.0	30.0	20.0	29.8	40	7.00	800	75.0	90.0	30.0	50.1
18	7.00	500	75.0	90.0	20.0	36.0	41	7.00	500	75.0	60.0	30.0	24.6
19	7.00	500	75.0	30.0	40.0	32.3	42	7.00	500	75.0	60.0	30.0	35.3
20	7.00	500	75.0	90.0	40.0	38.5	43	7.00	500	75.0	60.0	30.0	34.9
21	7.00	200	50.0	60.0	30.0	23.5	44	7.00	500	75.0	60.0	30.0	33.5
22	7.00	800	50.0	60.0	30.0	51.4	45	7.00	500	75.0	60.0	30.0	35.7
23	7.00	200	100	60.0	30.0	19.1	46	7.00	500	75.0	60.0	30.0	34.1

to understand the effects of variables better. As a response, it was employed the chemical oxygen demand (COD) expressed in mg L^{-1} and the percentage of removal of the antibiotic from aqueous solutions using MCM-41 adsorbent.

2.5. Adsorption experiments

Aliquots of 20.0 mL of 50.0–100.0 mg L^{-1} of the cephalexin at adjusted pH (3.00–11.0), were introduced in 50.0 mL conical flask containing MCM-41 adsorbent doses (200–800 mg L^{-1}) and put in an orbital shaker with temperature control for 30.0–90.0 min. After this time, the samples were taken with clean syringe filters, and the remaining cephalexin antibiotic was spectrophotometrically determined at 258 nm using LUV-100 Spectrophotometer [9]. The percentage of removal of the antibiotic was expressed by the Eq. (2) as follows:

$$\%\ \text{Removal} = 100.\frac{(C_o - C_f)}{C_o} \quad (2)$$

where C_o is the initial pharmaceutical concentration (mg L^{-1}). C_f is the final pharmaceutical concentration (mg L^{-1}).

3. Results and discussion

3.1. FTIR spectra

After preparing MCM41 and removing the surfactant from its structure by calcination at 550°C, the release of the organic matter from silica matrix was controlled by FTIR analysis of the MCM-41 (see Fig. 1). In the Fig. 1(a) two peaks at 2,914 and 2,848 cm^{-1} are observed corresponding to asymmetric and symmetric C-H stretching, respectively [46,47]. These two C-H bands are not displayed in Fig. 1(b) because of the releases of organic matrix in the calcination step to obtain MCM-41 silica. The bands in 1,221 and 1,056 cm^{-1} (Fig. 1(a)) and shoulder at 1,256 cm^{-1} and a band in 1,075 cm^{-1} (Fig 1(b)) confirms the formation of Si-O-Si bonds in the MCM-41 structure [46,47]. In addition, the bands at 3,410 cm^{-1} (Fig. 1(a)) and 3,442 cm^{-1} (Fig. 1(b)) could be assigned to Si-OH bands

[46,47]. The bends at 1,665 cm^{-1} (Fig. 1(a)) and 1,656 cm^{-1} (Fig. 1(b)) corresponding to bending of H-O-H of water molecules intercalated in the MCM-41 mesoporous silica [33]. The band at 1,494 cm^{-1} (Fig. 1(a)) is attributed to $^{+}N\text{-}(C)_4$ of CTAB that vanished after the calcination of the sample.

3.2. Analysis of X-ray diffraction

X-ray diffraction pattern analysis (Fig. 2) confirms the hexagonal structure in MCM-41 synthesised and especially the diffraction associated with the plate (100) at the low angle 2θ.

The peaks of the planes (100), (110) and (200) in the sample pattern of MCM-41 are well-suited to the patterns provided by other researchers [48].

Crystalline size of MCM-41 was calculated using the Debye–Scherer formula and its mathematical relation are shown in Eq. (3). In this equation D, β, λ and θ are crystalline sizes, the full width at half maximum of the peak corresponding to the plane, the wavelength of XRD radiation and angle is obtained from the 2θ value corresponding to XRD pattern, respectively.

$$D = \frac{0.94\lambda}{\beta\cos\theta} \tag{3}$$

According to the Debye–Scherer, the mean crystallite size of MCM-41 was 75.0 nm.

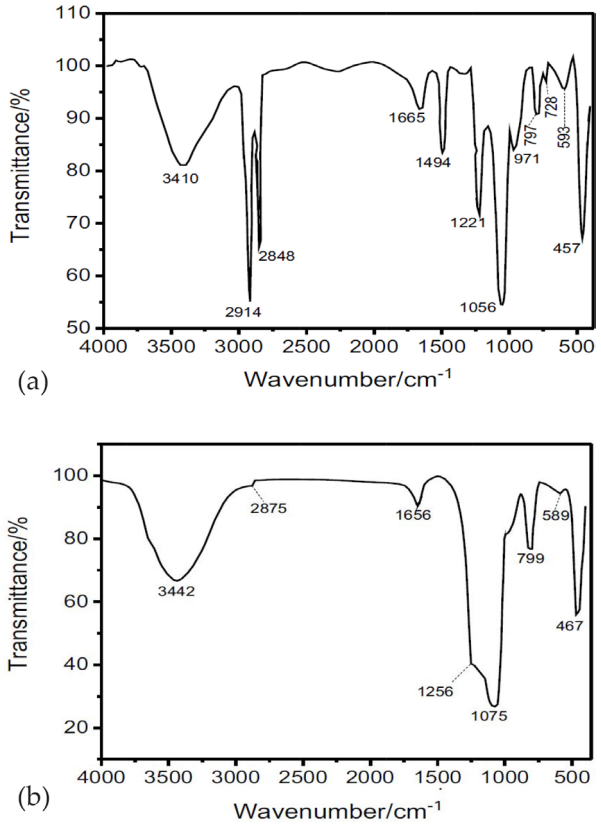

Fig. 1. FT-IR of MCM-41 (a) before Calcination and (b) after Calcination.

3.3. BET surface area

This analysis was carried out by adsorbed $N_{2(g)}$ at the level of mesoporous MCM-41 using a nitrogen analyser. The results of BET analysis are shown in Fig. 3. According to this analysis, the synthesised MCM-41 material presented pores with a diameter >2.0 nm (mesoporous material) and a surface area of 1,097 m^2g^1.

3.4. Analysis of FE-SEM

In the study, to investigate the surface morphology of synthesised MCM-41 mesoporous silica material and confirmation the adsorption of pollutants on the surface of mesoporous, the analysis FE-SEM were used. FE-SEM figure attributed to the before and after the adsorption process is shown in Fig. 4. According to Fig. 4(a), the surface of the adsorbent before the adsorption presented the high amount of adsorption sites. After the adsorption (Fig. 4(b)) the pollutants fill the sites of the adsorbent and empty the spaces on the surface of the adsorbent.

3.5. Statistical findings

In the selection of the model, all factors were checked, and the best model was selected according to the obtained *p*-value less than 0.05 that is significant at 95% of probability. A quadratic model, based on the Box-Behnken response surface methodology, was used to predict the percentage of removal of cephalexin from aqueous solution using the batch adsorption process. This equation is given as follows (see Table 2):

$$\text{Cephalexin removal} = 33.0 - 25.3A + 11.6B - 5.61C + 2.45D + 4.89A^2 \tag{4}$$

The units of these parameters are coded, where *A* is the pH; *B* is the adsorbent dosage (mg L^{-1}); *C* is the initial antibiotic concentration (mg L^{-1}); D is the temperature (°C).

Fig. 5 presents the normal plot of the residues for the response percentage of removal. As observed, this graph shows that the residuals follow a normal distribution pattern. Also, in Table 3 the ANOVA analysis for the percentage of removal of the antibiotic from aqueous solutions is presented. In the analysis of variance, if the *p*-value is less

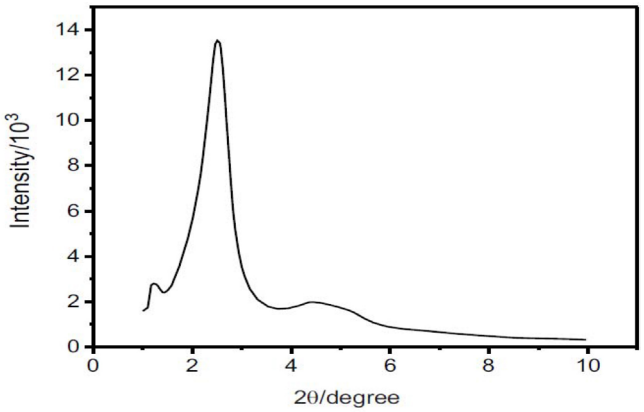

Fig. 2. Analyze of XRD.

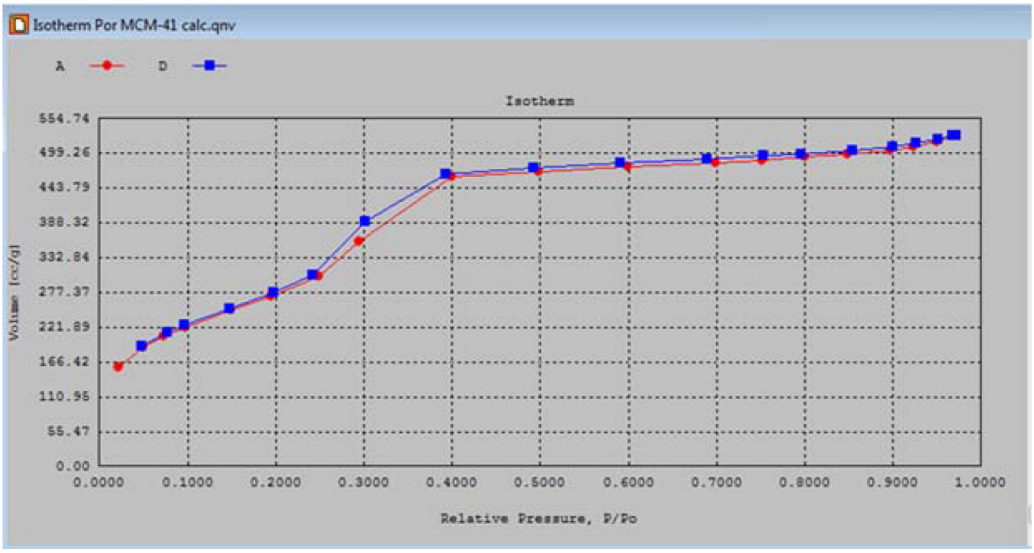

Fig. 3. Analyze of BET.

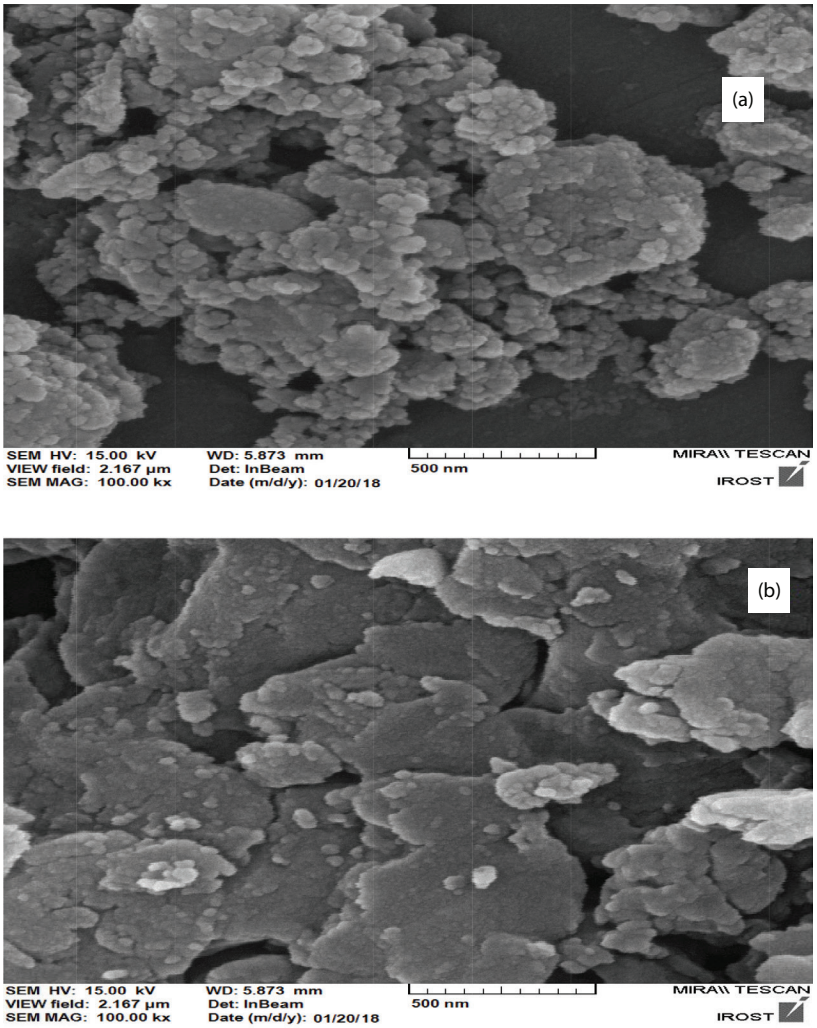

Fig. 4. FE-SEM images. (a) before adsorption and (b) after adsorption.

Table 2
Response surface regression: %Rem vs. pH; dosage; C_o; T; time

Term	Coefficient	Standard error of coefficient	*t*-Value	*P*-Value
Constant	33.0	1.44	23.0	0.000
pH	−25.3	0.88	−28.7	0.000
Dosage	11.6	0.88	13.2	0.000
C_0	−5.6	0.88	−6.38	0.000
Temp	2.45	0.88	2.79	0.010
Time	1.11	0.88	1.26	0.22
pH²	4.89	1.19	4.11	0.000
Dosage²	0.93	1.19	0.78	0.44
C_0^2	0.51	1.19	0.43	0.67
Temp²	0.80	1.19	0.67	0.51
Time²	0.81	1.19	0.68	0.50
pH × dosage	−2.49	1.76	−1.41	0.17
pH × C_0	2.08	1.76	1.18	0.25
pH × Temp	−1.23	1.76	−0.70	0.49
pH × time	−0.51	1.76	−0.29	0.77
dosage × C_0	−0.45	1.76	−0.25	0.80
dosage × Temp	0.43	1.76	0.24	0.81
dosage × time	0.13	1.76	0.07	0.94
C_0 × Temp	−0.13	1.76	−0.07	0.94
C_0 × time	−0.64	1.76	−0.36	0.72
Temp × time	−0.02	1.76	−0.01	0.99

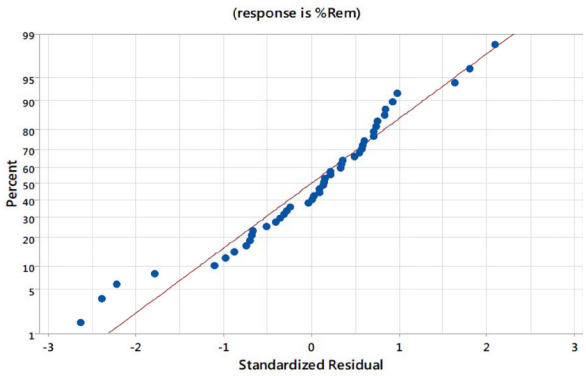

Fig. 5. Normal plot of the residues for the response percentage of removal.

than 0.05, or lack of fit is higher than 0.05, it represents that the model is significant. According to the table of variance analysis, the *p*-value and lack of fit were < 0.0001 and 0.798 respectively; therefore, the statistical model for the removal of cephalexin is significant. On the other hand, according to Table 3, it was observed that the parameter (E) contact time of the adsorbate solution with the adsorbent was not significant. The percentage of contribution of each factor is also shown in Table 3.

3.6. Effect of parameters

The primary parameter for optimisation of the maximum removal is the pH of the antibiotic solution (75.3% of the overall responses). The negative value of this parameter in Table 2 indicates that the increase in pH leads to a decrease in the removal percentage of cephalexin. According to Figs. 6 and 7, the increase in pH leads to the reduction of the percentage of cephalexin removal. Therefore, better removal of this antibiotic should be preferable in low pH values. The species of cephalexin at pH 3.00, 7.00 and 11.0 are presented in Fig. 6. At pH 3.00, the amino group is protonated that generates a positive charge on the antibiotic. The negative charge of MCM-41 electrically attracts the positive charge of the antibiotic at pH 3.00 since the pH_{zpc} is 2.29 [36]. Therefore, at pH 3.00, MCM-41 presents a superficial negative charge that attracts the cephalexin species at 3.0. In a study previously reported [9], the influence of pH on cephalexin adsorption onto SBA-15 mesoporous silica showed that according to the different species of cephalexin in different pH values and pH_{zpc} of adsorbent, the best adsorption efficiency was also obtained at low pH values [9].

The second parameter in order of importance on the overall response was the dose of adsorbent (15.9% of overall response). As its coefficient is positive, an increase in the adsorbent dosage, led to an increase in the percentage of removal of the antibiotic from aqueous solutions. The effect of adsorbent dose on the adsorption process is also shown in Figs. 6 and 7; the increasing of adsorbent dose leads to an increase in antibiotic adsorption. This behaviour is also expected as it was early reported [9,36] and also in the research conduct previously [9,36,49] that studied adsorption dyes and organic pollutant onto MCM-41 adsorbent, it was observed that the increase of MCM dosage resulted in increased contaminant removal [1,49]. The percentage of removal usually increases with the increase of adsorbent dosage to a limit of saturation of the adsorbent [1].

Table 3
Response surface regression: %Rem vs. pH; dosage; C_o; T; time

Source	DF	Seq SS	Contribution	Adj SS	Adj MS	*F*-Value	*P*-Value
Model	20	13,256	97.7%	13,256	663	53.64	0.000
Linear	5	12,986	95.7%	12,986	12,986	210	0.000
pH	1	10,209	75.3%	10,210	10,210	826	0.000
Dosage	1	2,158	15.9%	2,156	2,158	175	0.000
C_0	1	503	3.71%	503	503	40.7	0.000
Temp	1	96.0	0.71%	96.0	96.0	7.77	0.010
Time	1	19.7	0.15%	19.7	19.7	1.60	0.218
Square	5	217	1.60%	217	43.5	3.52	0.015
pH^2	1	205	1.51%	207	209	16.9	0.000
Dosage^2	1	3.20	0.02%	7.50	7.50	0.61	0.442
C_0^2	1	0.30	0.00%	2.30	2.30	0.18	0.671
Temp^2	1	3.10	0.02%	5.50	5.50	0.45	0.510
Time^2	1	5.70	0.04%	5.70	5.70	0.46	0.503
2-Way interaction	10	52.5	0.39%	52.5	5.20	0.42	0.921
pH × dosage	1	24.7	0.18%	24.7	24.7	2.00	0.170
pH × C_0	1	17.3	0.13%	17.3	17.3	1.40	0.247
pH × Temp	1	6.10	0.04%	6.10	6.10	0.49	0.490
pH × time	1	1.00	0.01%	1.00	1.00	0.08	0.774
Dosage × C_0	1	0.80	0.01%	0.80	0.80	0.06	0.802
Dosage × Temp	1	0.70	0.01%	0.70	0.70	0.06	0.811
Dosage × time	1	0.10	0.00%	0.10	0.10	0.01	0.943
C_0 × Temp	1	0.10	0.00%	0.10	0.10	0.01	0.943
C_0 × time	1	1.60	0.01%	1.60	1.60	0.13	0.720
Temp × time	1	0.00	0.00%	0.00	0.00	0.00	0.990
Error	25	309	2.28%	309	12.4		
Lack of fit	20	220	1.62%	220	11.0	0.62	0.798
Pure error	5	88.8	0.65	88.8	17.8		
Total	45	13,565	100%				
Standard deviation	R^2	R^2_{adj}	R^2_{pred}				
3.51	0.98	0.96	0.92				

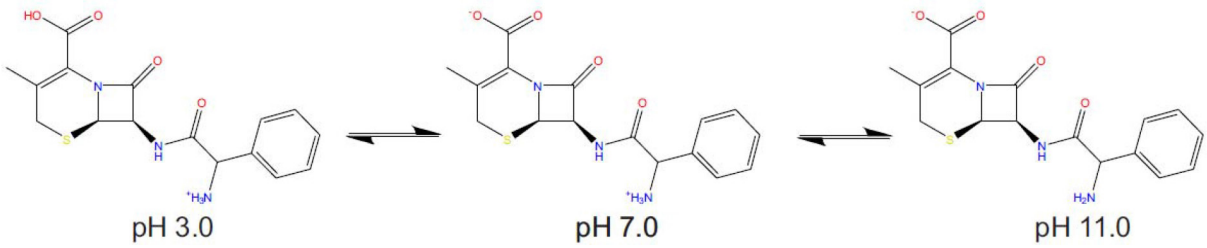

Fig. 6. Species of cephalexin at different pH.

The third parameter in order of importance on the overall response was the initial concentration of the antibiotic (C_0; 3.71% of overall response). As its coefficient is negative, it means that an increase in the initial antibiotic concentration will lead to a diminishing of the % removal, as early was observed [36,49–51]. Also, according to Figs. 6 and 7, by increasing initial antibiotic concentration, the efficiency of the adsorption process is decreased. In previous studies, [50,51] it was investigated the adsorption of benzene and ethanol on MCM-41 material where similar results were reported [50,51].

The fourth parameter in order of importance on the overall response was the quadratic pH value (1.51% of the overall response). This quadratic term was even more significant than the temperature parameter (0.71% of overall response). Only the achievements of these results already justify the Box-Behnken response surface methodology employed. It is possible to observe that the quadratic term of pH would be more significant to the overall optimization on the percentage of removal of the cephalexin antibiotic than the temperature, using univariate optimisation of the parameters. The positive coefficient of the

quadratic pH and temperature shows that increases in its values lead to increases in the percentage of removal.

The plot of predicted values vs. actual values of percentage of removal is presented in Fig. 7. This plot presents an intercept of 1.04 and a slope of 0.97, and R^2_{adj} of 0.97, indicating that the predicted value is close to the actual percentage of removal using MCM-41 as an adsorbent.

Fig. 8 presents the contour plot of all the parameters studied in this RSM. The arrows indicate the pathway to obtain the maximum percentage of removal of cephalexin antibiotic from aqueous solution, using MCM-41 adsorbent. These results indicate that the maximum removal would occur at low pH values, maximum adsorbent dosage, the minimum initial concentration of the adsorbate, higher temperature, and negligible effect of the time of contact between the adsorbent and adsorbate.

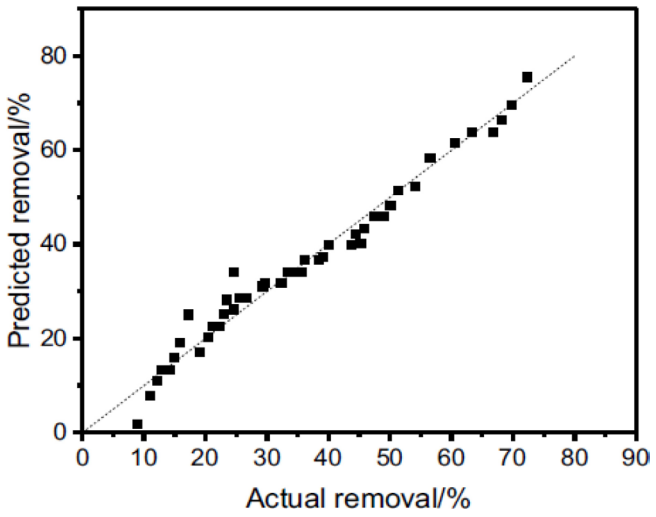

Fig. 7. Plot of predicted values vs. actual values of percentage of removal.

Fig. 9 presents the surface plot for a percentage of removal of cephalexin antibiotic from aqueous solution using MCM-41 mesoporous silica as an adsorbent. Observing the surface plots for the significant factors, again these results confirm the results of surface contour depicted in Fig. 9.

3.7. Optimisation of the adsorption process

In the optimisation process, the aim is to find a combination of levels of variables that maximally eliminate antibiotic cephalexin (Fig. 10). The response surface methodology selects and predicts the best operating mode within the range of applied variables. The model predicted about 90.3% of cephalexin antibiotic removal under optimal conditions, and the desirability factor was expressed 1.00 for these conditions (Fig. 10). The optimal conditions of the different adsorbent for adsorption of cephalexin antibiotics are summarised in Table 4 [10,52–54].

4. Conclusion

Analysis of FTIR represented the Si-OH, H-O-H and Si-O-Si bonds are formed in the structure of MCM-41. According to BET surface area, MCM-41 have pores with a diameter of more

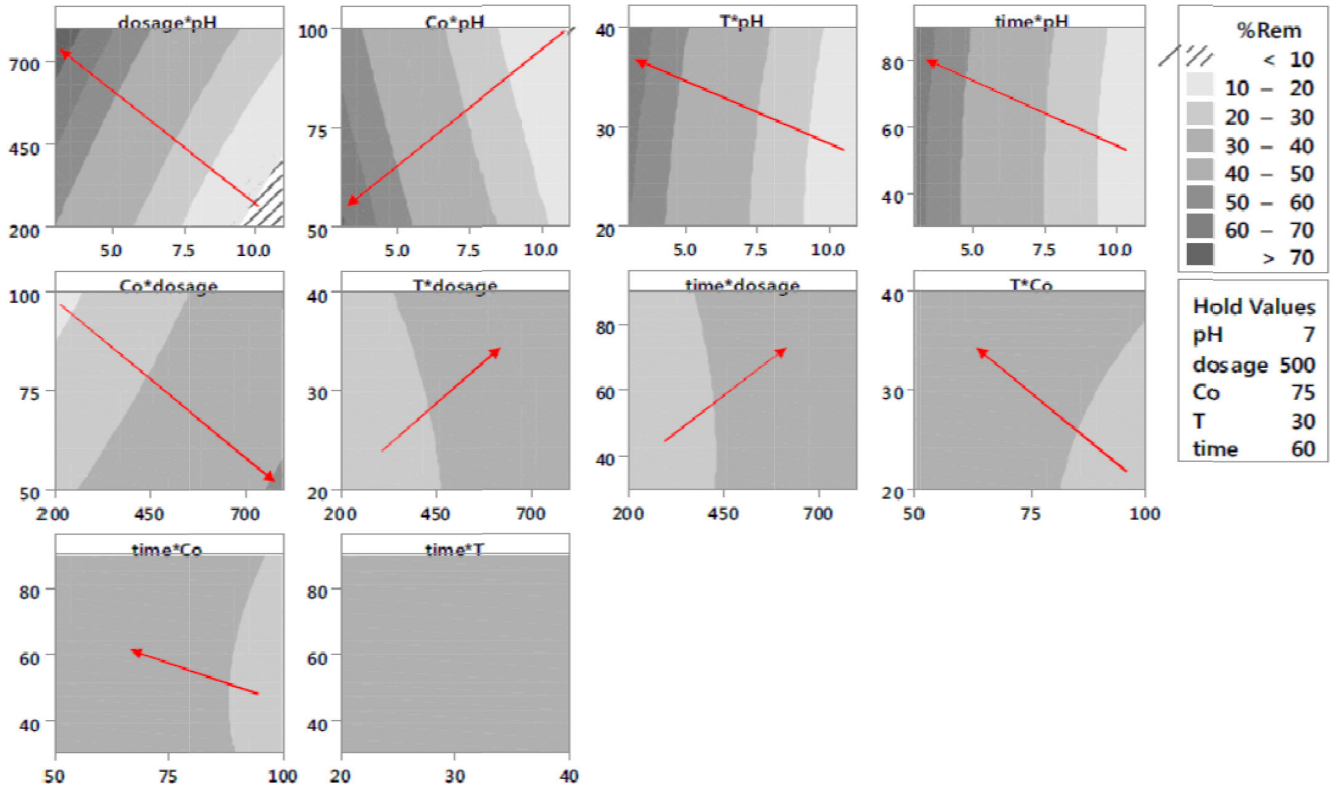

Fig. 8. The contour plot of all the parameters.

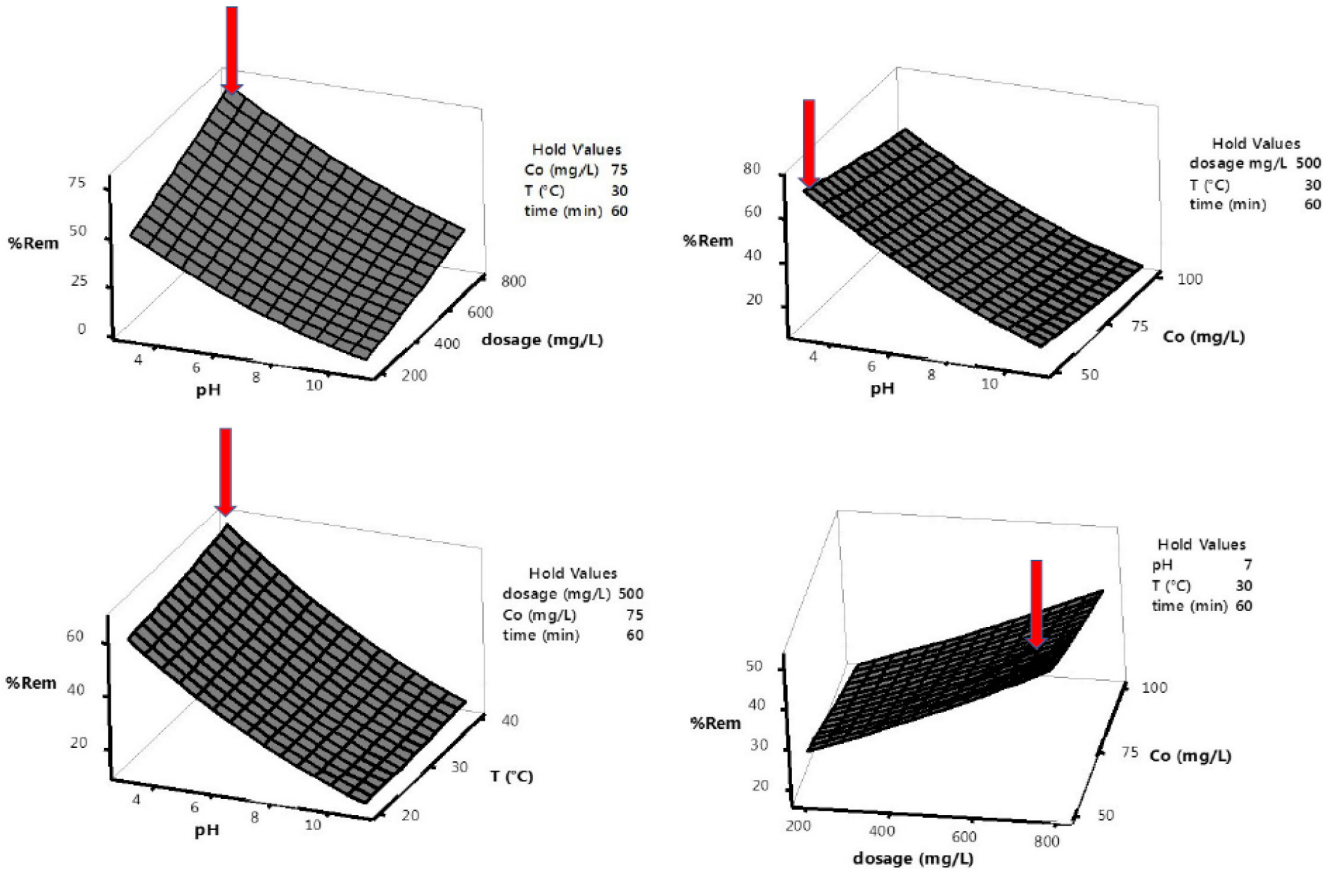

Fig. 9. Surface plot for a percentage of removal of cephalexin antibiotic.

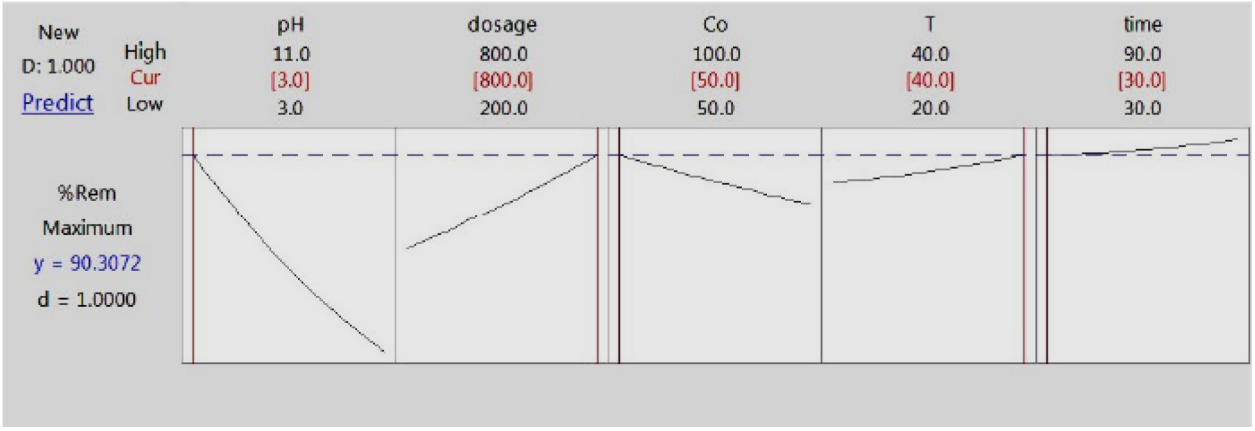

Fig. 10. Optimum parameters obtained by RSM.

than 2 nm and surface area of 1,097 m^2g^1 as well as the XRD spectra revealed the mean crystallite size of MCM-41 was 75.0 nm. The results of this study showed that the quadratic model was suitable for optimising the conditions for the optimal removal of cephalexin antibiotic using MCM-41 mesoporous silica as an adsorbent. It was tested a Box-Behnken response surface methodology, where the variables studied were initial pH of the adsorbate solution, adsorbent dosage, initial concentration of the antibiotic, temperature and time of contact between the adsorbent and adsorbate. According to statistical analysis, for the removal of maximum cephalexin antibiotic in optimal conditions, the values of the parameters were pH 3.00, an MCM-41 dose of 800 mg L^{-1}, the initial concentration of antibiotic of 50.0 mg L^{-1}, the temperature of 40.0°C and a contact time between the adsorbent and adsorbate of 30.0 min. Under these conditions, the efficiency is 90.3%, and the desirability factor is 1.00, using these experimental conditions. Therefore, according to the obtained results, the mesoporous silica can be used to eliminate the cephalexin antibiotic in optimal conditions designed by RSM.

Table 4
Optimal conditions of different absorbent for adsorption of cephalexin antibiotics

Optimum conditions and adsorption rate of cefalexin antibiotics	Adsorbent
Maximum removal of cephalexin antibiotic in optimal conditions was at the values of the parameters were pH 3.00, an MCM-41 dose of 800 mg L^{-1}, the initial concentration of antibiotic of 50.0 mg L^{-1}, the temperature of 40.0°C and a contact time between the adsorbent and adsorbate of 30.0 min	MCM-41
The maximum adsorption capacity was obtained 233 mg/g based on the Langmuir model [46]	Walnut shell-based activated carbon
It was observed that the maximum adsorption capacity was 270 $mg.g^{-1}$ [47]	Modified Walnut Shell
The Langmuir isotherm was suitable for correlation of equilibrium data with the maximum adsorption capacity of 48.7 mg/g [48]	Activated carbon (PPAC) derived from pomegranate peel
The adsorption intensity was found to be increased as the aqueous phase pH increased, and had a maximum at pH = 6.1 [49]	Bentonite and activated carbon
The maximum CEX removal efficiency was 28.0% and 89.0% for NZ (natural zeolite) and CZ (zeolite coated with manganese oxide nanoparticles), respectively, at pH 7 [10]	Natural zeolite and zeolite coated with manganese oxide nanoparticles

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Ethical issues

The author hereby credential that all data collected during the Research is as expressed in the manuscript, and no data from the study has been or will be published separately elsewhere.

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Supplementary material

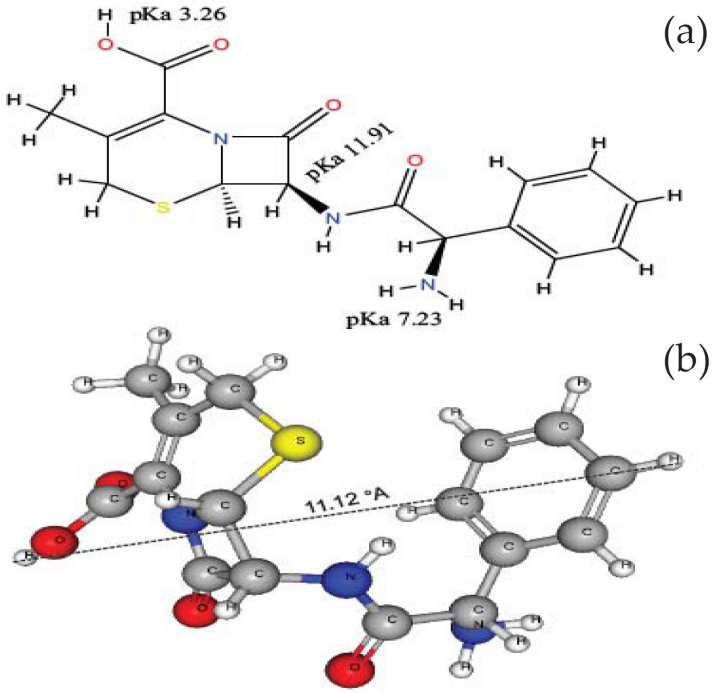

Fig. S1. (a) Structural formula of cephalexin (347.389 g mol^{-1}, $C_{16}H_{17}N_3O_4S$ CAS 15686-71-2) pK_a values were given; (b) optimized three-dimensional structural formula of cephalexin. The dimensions of the chemical molecule and the physical and chemical properties were calculated using MarvinSketch version 18.15.0. Van der Walls volume 290.74 A^3; Van der Waals surface area 435.11 A^2 (pH 7.0); Polar surface area 142.48 A^2 (pH 7.0); Dipole Moment 6.98 Debye; Log P 0.65; Solubility 0.297 g L^{-1}.