RESPONSE SURFACE OPTIMIZATION AND MODELING OF AMMONIUM CHLORIDE ACTIVATION PROCESS OF BENTONITE

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RESUME

Ce travail a consisté en l'optimisation et la modélisation de l'activation d'une bentonite avec le chlorure d'ammonium en utilisant la méthode des plans d'expériences, et en particulier la méthodologie de surface de réponse (RSM). Un plan central composite orthogonal (CCOD), a permis la détermination de l'influence des effets simultanés et de l'interaction des paramètres opératoires sur la surface spécifique (S_{BET}). Les paramètres étudiés sont la température de l'activation (50-90°C), le temps de contact (2-6h), le rapport massique liquide/solide (4-7g/g) et la concentration de la solution d'activation (1-2 mol L^{-1}).

Les valeurs optimales des paramètres d'activation donnant une surface spécifique maximale ont pu être déterminées (température : 98.28°C, temps du contact : 6.828 h, rapport massique liquide/solide : 4.627 g/g, concentration de la solution d'activation : 0.793 mol L^{-1}). La surface spécifique de l'argile activée sous ses conditions optimales étant de 87.545 m^2g^{-1} et elle est en bonne adéquation avec la valeur prédite par le modèle (89.9 m^2g^{-1}). Le modèle de second ordre obtenu pour la surface spécifique de la bentonite activée est en bonne adéquation avec les résultats expérimentaux. Le traitement d'une bentonite tunisien avec une solution de chlorure d'ammonium provoque une augmentation de la surface spécifique de 47.3 à 87.545 $m^2\ g^{-1}$ correspondant à une augmentation de 85%.

ملخص

هذا العمل يتمحور حول إيجاد التصميم الأمثل و الظروف المثلى لعملية تنشيط طين باستخدام محلول كلوريد الأمونيوم و ذلك بعد نمذجة عملية التنشيط باستعمال هندسة التجارب وفق طريقة منهجية استجابة السطح (RSM).

استخدم مخطط عاملي ذات مستويان (2k) لدراسة أربعة عوامل رئيسية مؤثرة في عملية التنشيط بالإضافة إلى بعض العلاقات المتداخلة بين العوامل و ذلك بقياس المساحة السطحية النوعية للطين (قيمة الاستجابة)، و هي تأثيرات درجة الحرارة ، زمن التفاعل، النسبة الكتلية سائل /صلب و تركيز محلول كلوريد الأمونيوم .

تم تحديد الشروط المثلى للعوامل الاساسية المؤثرة و المؤدية إلى أكبر مساحة سطحية نوعية فكانت النتائج كالتّالي (درجة الحرارة :98,828 درجة مؤوية ، مدة التفاعل: 6,828 ساعة، النسبة الكتلية سائل /صلب: 4,627 غ/غ ، و تركيز محلول كلوريد الأمونيوم 0,793 (mol/L^{-1}) و التّي مكّنت من الحصول على مساحة سطحية نوعية 87,545 م^2/ غ حيث كانت قبل التنشيط 47,3 م2/ غ . كما كانت النتائج تشير إلى أنّ نموذج المحاكاة لعملية التنشيط المتحصل عليه يلائم نتائج التجارب التي أجريت (المساحة السطحية النوعية المتحصل عليها من قبل نموذج المحاكاة 89,9 م^2/ غ).

و عليه نخلص إلى أنّ عملية تنشيط الطين المستخدم باستعمال محلول كلوريد الأمونيوم مكّن من التّرفيع في مساحته السطحية النوعية بما يعادل نسبة 85 % .

الكلمات المفاتيح: تنشيط الطين ، منهجية استجابة السطح (RSM) تحليل التغير (ANOVA) ، أمثلة .

ABSTRACT

In this work, a bentonite activation processes with ammonium chloride were investigated using the Response Surface Methodology (RSM). The specific surface areas (S_{BET}) of the treated samples were chosen as the process response. The effect of four independent factors; namely activation temperature (50-90°C), contact time (2-6h), liquid-to-solid mass ratio (4-7g/g) and ammonium chloride concentration (1-2 mol L^{-1}) on the process response were determined. A basic experimental design has been done according to a Central Composite Orthogonal Design matrix (CCOD). Indeed, it aimed at obtaining a relationship between the specific surface area and the factors. The optimum conditions for ammonium activation process which maximized the specific surface area (S_{BET}) were found to be 98.28°C, 6.828 h, 4.627 g/g and 0.793 mol L^{-1} respectively for temperature, contact time, liquid-to-solid mass ratio and ammonium chloride concentration. Under these optimum conditions a specific surface area of activated bentonite of 89.9 m² g^{-1} was predicted. The experimental value obtained under optimum conditions (87.545 m² g^{-1}) was in good agreement with the predicted value. The second order polynomial model developed was satisfactory in fitting the experimental results. The treatment of Tunisian bentonite with ammonium chloride solution produces an increase in the specific surface area from 47.3 to 87.545 m² g^{-1} corresponding to an 85% increase.

KEYWORDS: Activated bentonite; response surface methodology; ANOVA; optimization.

1 INTRODUCTION

Bentonites which are available in large quantities and at low price, are of great interest in drug manufacturing and pollution control [1, 2]. Bentonites were used as adsorbents, to remove not only dyes but also other organic and inorganic pollutants such us heavy metal [3, 4, 5], humic substances [6, 7], phenol [8,9] and toluene [10]. The future of clay science looks bright, exciting, and promising [1].

In order to improve the adsorption capacities of bentonites, various chemical or physical processes have been applied. These processes modify the physical and chemical properties of clay by obtaining a partly dissolved material of increased specific surface area, surface acidity and porosity [11,12]. The most widely used processes are acid activation and cation exchange [1].

In general the methodologies adopted to carry out these processes were classified in two categories [13,14]. In the classical experimental method, one factor was varied at a time, while all the others were kept constant [14]. In addition to being lengthy and expensive, the classical method could not evaluate the interaction between the variables and it was less effective [15]. The weaknesses and limitations of the classical method have been overcome by the use of Response Surface methodology (RSM). Unlike the classical method, the RSM is a collection of statistical techniques which use the experimental design in which the effects of more than one factor on response are evaluated. Indeed, This approach is a powerful technique that has several advantages [16] : To begin with, it considerably reduces the number of experiments. Next it made possible to evaluate the interaction between factors. Moreover, it finds the best fitting mathematical model and determines the optimum conditions of the process.

The aim of this study is to evaluate the optimum conditions for the ammonium chloride process activation of bentonite from Gabes (Tunisia). The specific surface area of activated bentonite was used as the response function. A Central Composite Orthogonal Design (CCOD) of Response Surface Methodology (RSM) was applied in this work to examine the influence of the following four relevant factors on the specific surface area: temperature, contact time, solid-to-liquid mass ratio and acid concentration.

2 MATERIALS AND METHODS

2.1 Materials

The raw bentonite used comes from a quarry located in El-Hamma (Gabes, Tunisia). The chemical analysis of the raw material is shown in Table 1.

Table 01: Chemical composition of raw bentonite (wt %)

Compounds	SiO_2	Al_2O3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	SO_3	LOI*	Total
Value	45.59	18.69	12.5	1.99	3.74	0.72	1.01	0.52	15.31	99.39

* loss on ignition (the sample is heated for 2 hours at 1000°C)

2.2 Ammonium activation

The activation process was carried out in jacketed glass reactor. A specified mass of raw bentonite was dispersed in 500 ml of the ammonium chloride solution. The suspension was stirred at a specified temperature. On completion of the specified activation time, the dispersion was filtered and the activated bentonite was washed with ultra-pure water until it was free of Cl^- ions, as tested by $AgNO_3$, and dried for 24 hours. Then the material was crushed and screened through a 100 μm sieve. The specific surface areas (S_{BET}) of the ammonium activated bentonites samples were measured by nitrogen sorption.

2.3 Sample Characterizations

The specific surface area and pore volume of bentonite samples were measured by nitrogen sorption using a Quantachrome Autosorb Automated Gas Sorption System, after degassing the samples at 120°C for 24 h and under a residual pressure of 1 mTorr. The specific surface area was calculated by the Brunauer–Emmet-Teller (BET) method and the pore size distribution was calculated using the Barret-Joyner-Halenda method (BJH). Fourier Transform Infrared (FTIR) spectra of the raw and ammonium activated bentonite were recorded using a Spectrum 400 Perkin Elmer operating in the range of 400-4000 cm^{-1} at a resolution of 4 cm-1. The XPS spectra were recorded in a Thermo VG ESCALAB 250 instrument using monochromated Al Kα radiation (1486.6 eV).

2.4 Design of experiments

In order to determine the optimum conditions for bentonite activation process by the ammonium chloride solution, the response surface methodology was used. This method is extremely useful for developing, improving, and optimizing processes [17, 18].

Four independent activation parameters were selected in this study, the activation temperature T (Z_1), the contact time t (Z_2), the liquid-to-solid mass ratio R (Z_3) and the concentration of ammonium chloride solution C (Z_4). Other parameters such as agitation speed were kept constant (550rpm). The specific surface area of activated bentonite (S_{BET}) was used as response function.

In the present study a Central Composite Orthogonal Design (CCOD) for four parameters was implemented. Tow levels of variation for each variable were considered. Each of the chosen parameters (Z_i) was coded at levels: -α, -1, 0, +1 and +α according to the following Equation [19]:

$$X_i = \frac{Z_i - Z_0}{\Delta Z} \qquad (1)$$

Where X_i is the dimensionless coded value of the variable Z_i, Z_0 the value of the Z_i at the centre point, and ΔZ the step change value. The relationship between the parameters and response surface was explained by the following quadratic equation:

$$Y = b_0 + \sum_{i=1}^{k} b_i X_i + \sum_{i=1}^{k} b_{ii} X_i^2 + \sum_{i=1}^{k-1} \sum_{j=i+1}^{k} b_{ij} X_i X_j + \varepsilon \qquad (2)$$

Where, Y is the predicted response, b_0 the intercept term, b_i the linear effect, b_{ii} the squared effect, b_{ij} the interaction effect and ε is a random error [19, 20].

The CCOD, used in this study to fit the second-order polynomial model, was obtained by upgrading the full factorial (2^4) designs with one additional point in the experimental center and the corresponding number of "starlike" points (α and -α levels) [19]. The experimental ranges and levels investigated in this work are given in Table 2. The response of the experiment at the experimental center is the mean of four experiments.

Table 02: Experimental ranges and levels of independent variables

	Factor	*Range and levels*					*Variation intervals ΔZ*
		-1.414	-1	0	1	+1.414	
X_1	Temperature :T (°C)	41.72	50	70	90	98.280	20
X_2	Contact time: t (h)	1.172	2	4	6	6.828	2
X_3	Liquid to solid mass ratio :R (g g^{-1})	3.379	4	5.5	7	7.621	1.5
X_4	Concentration of NH_4Cl : C (mol L^{-1})	0.793	1	1.5	2	2.207	0.5

The value of α is determined by the equation (3) [21], where N_F and N_T are respectively the numbers of the full factorial designs and the Central Composite Orthogonal Design:

$$\alpha = \left[\frac{N_F \left(\sqrt{N_T} - \sqrt{N_F} \right)^2}{4} \right]^{\frac{1}{4}} \qquad (3)$$

The design matrix together with the outcomes of design points and the predicted values are shown in Table 3. The significant terms in the model were found through the analysis of variance (ANOVA) for each response. The optimum conditions of activation process were obtained by solving the regression model. In order to confirm the optimization result, run under the optimized predicted conditions were performed.

Table 03: Experimental design matrix for CCOD, observed and predicted responses

Run	*X_1*	*X_2*	*X_3*	*X_4*	*S_{BET} ($m^2 g^{-1}$)*		
					Observed	*Predicted*	*Residual*
1	-1	-1	-1	-1	35.73	38.82	-3,12
2	1	-1	-1	-1	69.30	65.91	3.39
3	-1	1	-1	-1	67.50	64.01	3.49
4	1	1	-1	-1	77.80	78.15	-0.35
5	-1	-1	1	-1	58.43	57.21	1.22
6	1	-1	1	-1	71.77	71.19	0.58
7	-1	1	1	-1	74.00	74.71	-0.71
8	1	1	1	-1	76.83	75.73	1.1
9	-1	-1	-1	1	55.33	53.80	1.53
10	1	-1	-1	1	72.29	71.86	0.43
11	-1	1	-1	1	67.53	68.39	-0.86
12	1	1	-1	1	74.91	73.50	1.41
13	-1	-1	1	1	71.24	72.19	-0.95
14	1	-1	1	1	75.25	77.14	-1.89

15	-1	1	1	1	77.30	79.09	-1.79
16	1	1	1	1	72.88	71.09	1.79
17	0	0	0	0	69.68	66.66	3.02
18	1.414	0	0	0	78.43	82.45	-4.02
19	-1.414	0	0	0	70.32	68.96	1.36
20	0	1.414	0	0	72.12	73.43	-1.31
21	0	-1.414	0	0	60.61	59.90	0.72
22	0	0	1.414	0	67.54	66.54	1.00
23	0	0	-1.414	0	51.58	55.24	-3.66
24	0	0	0	1.414	71.21	70.31	0.9
25	0	0	0	-1.414	59.69	63.01	-3.32

3 RESULTS AND DISCUSSIONS

3.1 Characterization

The pore size distribution of the raw and ammonium activated bentonite samples are shown in Fig.1. An increase in the average pore volume was observed after the activation process. The infrared spectra of the bentonite before and after ammonium activation, is shown in Fig. 2. In both spectra there is a peak at near 1000 cm^{-1} attributed to Si-O stretching vibration. A three bands in hydroxyl bending region 950-850 cm^{-1} assigned to Al2OH(near 919 cm^{-1}), AlFeOH(near 880 cm^{-1}) and AlMgOH(near 850 cm^{-1}). The Al_2OH stretching vibration frequency is observed at 3620 cm^{-1} [22, 23]. The quartz impurity is detected by the presence of the doublet at 799 and 779 cm^{-1}. For the activated bentonite, the bending and stretching vibrations of NH4+ are respectively observed at 1430 cm^{-1} and 3275 cm^{-1} (Fig.2b). The results given by the FTIR spectrum in our study is in accordance with those of authors who worked on NH_4^+ exchanged clay minerals [22, 24].

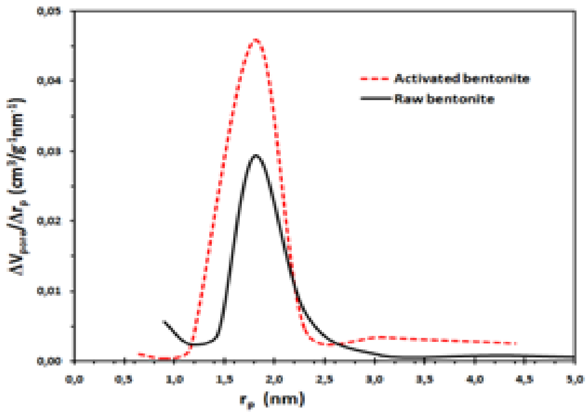

Figure 01: The pore size distribution of the raw and ammonium activated bentonite samples

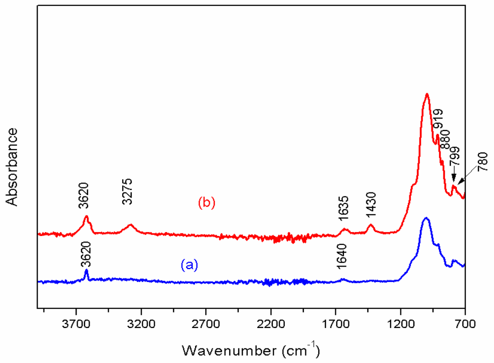

Figure 02: FTIR spectra of (a) raw bentonite and (b) ammonium activated bentonite

Fig. 3 displays the survey regions of the raw bentonite and the ammonium activated bentonite. The main peaks Al2p, Si2p, K2p, Ca2s, O1s and Na1s are centered at ~75, 103, 293-296, 347, 533, and 1072 eV, respectively. The minor C1s peaks (285 eV) are due to an unavoidable hydrocarbon surface contamination. Interestingly, one can clearly note the lack of N1s at 402 eV from the raw bentonite and its appearance after modification by ammonium chloride. Concomitantly, the intensity of the Na1s peak drastically decreases due to the modification of raw bentonite. The surface elemental composition is reported in Table 4.

Table 04: Apparent surface elemental composition of betonite before and after ammonium activation

Materials	*Al*	*Si*	*C*	*K*	*Ca*	*N*	*O*	*Fe*	*Na*	*Mg*
Raw bentonite	10.5	20.9	9.33	0.5	0.36	0	55.1	0.73	1.03	1.64
Ammonium activated bentonite	9.03	20.6	7.4	0.44	0.31	0.73	59.9	0.97	0.18	1.21

The K/Na atomic ratio increases from 0.49 to 2.44 due to the exchange of sodium by the ammonium cations. The cation exchanges affects sodium only as the potassium content does not change significantly. This is a true cation exchange as sodium is replaced by the ammonium cation without any adsorption of chlorides from NH_4Cl as testified by the absence of any Cl2p peak at ~198 eV, a binding energy position characteristic of chlorides. Another important observation concerns the aluminosilicate sheets: the Si/Al atomic ratio increases only by 10% from 2 to 2.3.

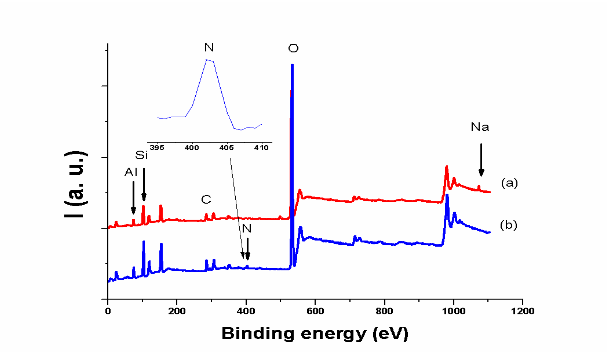

Figure 03: XPS survey regions of raw bentonite (a) and ammonium activated bentonite (b)

3.2 Fitting the regression equation

The analysis of variance (ANOVA) was applied in this study so us to fit the second order model equation to the experimental data. The Main effect, interaction effect, coefficient of the model, standard error and probability for the full CCOD are presented in Table 5. The significance of each obtained coefficient and the model equation were checked by the student's and Fisher's statistical tests (t-value, F-value) and the value of probability P > F (p-value > F) [25]. The Fisher's criterion F-value is the ration of mean square due to regression to the mean square due to real error. The regression coefficient with p-value less than 0.01 indicates that the model terms are high statistically significant [26].

Regression coefficient with p-value less than 0.05 indicates that the model terms are significant with 95% confidence level [19].

The ANOVA results (Tables 5 and 6) have clearly shown that the variables X_1, X_2 and X_3 have a high significant effect on bentonite activation process (p-value < 0.0001). Since they have a p-value < 0.05, the X_4 factor, the interactions X_1X_2, X_1X_3, X_1X_4, X_2X_3, X_2X_4 and square effects (X_1^2, X_3^2) are considered as significant factors.

In addition, the probability results (p-value > 0.05) have showed that only X_3X_4, X_2^2 and X_4^2 were statistically insignificant effect on the bentonite activation process. Based on the F-test, the regression model was with 95% confidence adequate since its calculated F-value was higher than the critical F-value. The fitness of the quadratic polynomial model was checked by the determination coefficient (R^2). In this study, the value of the correlation coefficient (R^2=0.95) reflected a strong relationship between the response (Specific Surface area of bentonite) and the activation parameters. In the same way the p-value (Prob. > F) less than 0.05 showed that the model was adequate with 95% confidence (Table 7).

Table 05: The ANOVA results for the regression coefficients before rejecting the insignificant coefficients

Model Term	*Coefficient Estimate*	*standard Error*	*t-value*	*p-value*
Constant	66.199	1.947	33.99	<.0001
temperature	4.772	0.726	6.57	<.0001
Temps	4.785	0.726	6.59	<.0001
Ratiot	3.994	0.726	5.5	0.0003
concentration	2.583	0.726	3.56	0.0052
temperature*time	-3.237	0.812	-3.99	0.0026
temperature*ratio	-3.278	0.812	-4.04	0.0024
time*ratio	-1.923	0.812	-2.37	0.0393
temperature*concentration	-2.257	0.812	-2.78	0.0194
Time*concentration	-2.649	0.812	-3.26	0.0085
ration*concentration	-0.256	0.812	-0.31	0.7593 ^{IS}
temperature*temperature	4.523	1.148	3.94	0.0028
Time*time	0.517	1.148	0.45	0.6619 ^{IS}
ratio*ratio	-2.886	1.148	-2.51	0.0307
concentration*concentration	0.060	1.148	0.05	0.9596 ^{IS}

^{IS} insignificant coefficient

Table 06: The ANOVA results for the regression coefficients after rejecting the insignificant coefficients

Terme	*Degrees of freedom*	*Sum of squares*	*Estimation*	*Erreur standard*	*F-value*	*t-value*	*p-value*
Constante	1		66.661	1.293		51.570	<.0001
Température	1	455.444	4.772	0.646	54.521	7.380	<.0001
Temps	1	457.810	4.785	0.646	54.805	7.400	<.0001
Rapport	1	319.040	3.994	0.646	38.192	6.180	<.0001
Concentration	1	133.442	2.583	0.646	15.974	4.000	0.002
Température*temps	1	167.638	-3.237	0.723	20.068	-4.480	0.001
Température*Rapport	1	171.938	-3.278	0.723	20.583	-4.540	0.001
temps*Rapport	1	59.175	-1.923	0.723	7.084	-2.660	0.020
Température*Concentration	1	81.496	-2.257	0.723	9.756	-3.120	0.008
temps*Concentration	1	112.307	-2.649	0.723	13.444	-3.670	0.003
Température*Température	1	163.646	4.524	1.022	19.590	4.430	0.001
Rapport*Rapport	1	66.613	-2.886	1.022	7.974	-2.820	0.014

Table 07: The ANOVA results for the quadratic polynomial model

Source of variations	*Degrees of freedom*	*Sum of squares*	*Mean square*	*F-value*	*Prob. > F*	*R carrée*
Modèle	11	2188.497	198.954	23.817	<.0001	0.953
Résidus	13	108.596	8.353			
Total	24	2297.092				

After rejecting the statistically insignificant regression coefficients, the second-order regression model obtained by application of RSM has the following form:

$$Y = 66.661 + 4.772X_1 + 4.785X_2 + 3.994X_3 + 2.583X_4 - 3.237X_1X_2 - 3.278X_1X_3 - 1.923X_2X_3 - 2.257X_1X_4 - 2.649X_2X_4 + 4.524X_1^2 - 2.886X_3^2 \quad (4)$$

This is an empirical relationship between specific surface area of ammonium activated bentonite (Y) and the test factors. In where X_1, X_2, X_3 and X_4 are respectively, the coded values for temperature of activation (T), contact time (t) , liquid-to-solid mass ratio (R) and concentration of ammonium chloride solution (C).

The ranking importance of each factor or interaction was given by the numerical value and sign of the regression-coefficient. A positive sign indicates that with an increase in factor value there is an increase in response value, whereas a negative sign means a decrease in response value [19].

From the model equation, it was apparent that the temperature X_1 and time X_2 were the most important variables for the activation process. The second important factor was the liquid-to-solid ratio X_3. The positives values of each one of their regression coefficients meant that an increase in their levels has brought about an increase in the surface area of activated bentonite. However all the double interactions have a negative sign. The second order of temperature has a high positive value and the second order of liquid to solid ratio has a negative value.

3.3 Assessment and verification of the optimum activation parameters

After developing the adequate model of second order response surface, the process was optimized for the response Y. Optimization was achieved to obtain the values of T, t, R and C, which maximized Y (S_{BET}). The results have showed that the maximum surface area of activated bentonite was reached at a temperature of 98.28°C, a contact time of 6.828 h, a liquid-to-solid mass ratio of 4.627 g/g and an ammonium chloride concentration of 0.793mol L^{-1}.

In order to verify the validity of this Optimization, an activated bentonite was prepared under the optimum conditions. The surface area obtained was in close agreement with the predicted value as shown in Table 8.

Table 08: Optimum conditions, predicted and experimental value of surface area of activated bentonite under those conditions

Optimum conditions				*Surface area of Activated bentonite*		
Temperature (°C)	Contact time (h)	Liquid to solid mass ratio (g/g)	Ammonium chloride concentration (mol/l)	Experimental	Predicted	Erreur (%)
98.28	6.828	4.627	0.793	87.5 ± 0.6	89.9 ± 3.4	2.6

The variance was calculated for the predicted response (89.9) by the equation 5; the standard deviation (3.4), is the square root of the variance.

$$VarY_P = \left[x_P\right]\left[{}^{t}XX\right]^{-1}\left[{}^{t}x_P\right]\sigma_{res}^{2} \qquad (5)$$

The standard deviation (0.6) of the experimental response was calculated from the four essays at the experimental center (70°C ; 4 h ; 5.5 g g^{-1} ; 1.5 mol L^{-1}).

3.4 Analysis of response surface plot

In order to better clarify the impact of each factor and its interaction on the activation process, a 3D response surface and 2D contour were plotted.

The simultaneous effect of contact time (t) and liquid-to-solid mass ratio (R) on the surface area of activated bentonite was showed in Fig. 4. The remaining factors were kept constant (T=98.3 °C, C=0.793 mol L^{-1}).

At a low level of R (4 g/g), SBET increased from 75.2 to 89.4 m² g^{-1} when t increased from 2 h to 6h50. Similarly, at a high level of R (7 g/g), SBET increased from 77.8 to 82.7 m² g^{-1} when t increased from 2 to 6h50.

At a high level of t (6h50), S_{BET} increased from 89.4 to 89.9 m² g^{-1} when R increased from 4 to 4,627 g/g and reaches a maximum value (89.9 m² g^{-1}), before it began to decrease to 82.7 m² g^{-1} when R reach the maximum (R= 7 g/g).

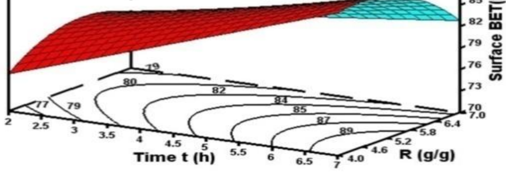

Figure 04: ***Response surface plot displaying the effects of the mutual interactions between contact time (t) and liquid to solid mass ratio (R), (T=98.28°C , C=0.793 mol/l)***

4 CONCLUSIONS

Ammonium chloride activation bentonite process was modulated and optimized using a Response Surface Methodology. The experimental sets have been designed according to full central composite orthogonal designs (CCOD) to obtain a second-order mathematical model. The statistical significance of each regression coefficient and the degree of fit of the model equation were evaluated by the analysis of variance (ANOVA) followed by Fisher’s and student’s statistical tests.

The effect of four factors was studied: activation temperature (T), contact time (t), liquid-to-solid mass ratio (R) and concentration of ammonium chloride solution (C). The specific surface area of activated bentonite was taken as the process response. The regression model developed was found adequate with 95% confidence. The determination coefficient of the results obtained was high (R^2=0.95), indicating a good fit of the model with the experimental data.

From the results of this work it could be deduced that the treatment of bentonite by ammonium chloride solution was controlled by the interaction of the four factors. The optimum values of the variables were found to be 98.28°C, 6.828h, 4.627 g/g and 0.793mol L^{-1}. The maximum specific surface area of activated bentonite was predicted to be 89.882 m² g^{-1}. The experimental response obtained under these optimum conditions was: 87.545 m² g^{-1}. The XRD results proved a partial destruction for the bentonite structure and pore size distribution was strongly affected by ammonium activation process.

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