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Modeling of the sorption of crude oil on a polyurethane foam-powdered activated carbon composite

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ABSTRACT

In an adsorption process, spontaneous accumulation of molecules takes place at the surface of solids as compared to the bulk phase. Adsorption is used for the recovery of undesirable components from a liquid mixture by establishing van der Waals bonds between the solid surface and the attracted components. Nowadays, adsorption is recognized as one of the major significant separation processes amid the physical, biological, and chemical processes. Activated carbon adsorption has been widely used for the purification of water and wastewater and for the treatment of numerous environmental problems. In this work, polyurethane (PU) foam-activated carbon composites were synthesized and characterized. The mixing of a number of well-defined ingredients produces flexible foam of alveolar structure after polymerization reactions. The prepared PU-powdered activated carbon composites were used to investigate the sorption of raw crude oil. Multiple experimental manipulations were combined to understand this behavior. The application of a design of experiments (DOE) method was used to model the adsorption kinetics.

Keywords: PU foams; Activated carbon; Modeling; Sorption; Crude oil; Design of experiments

1. Introduction

Oil spills which frequently invade the coasts are a real environmental scourge. They pollute oceans and destroy the marine fauna and flora entailing the degradation and scarcity of these valuable resources. Today, the challenge is to rapidly recover the spilled oil while minimizing the risk of pollution. An existing method is to use surfactants which reduce the oil–water interfacial tension, allowing the dilution and biodegradation of oil in water. However, the surfactants are expensive and contain toxic products [1]; the challenge is to find an environmentally friendly material with high sorption kinetics for heavy oils such as crude oil (petroleum). Several research works have shown that porous carbons based on active carbon and carbon nanotubes meet the criteria [2–5]. Other materials such as activated carbon-loaded foams can also be significantly interesting due to their simultaneous flexible and robust properties. The components absorbed by such foams can be extracted by simple compression [6].

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The sorption of crude oil by the polyurethane foam filled with powdered activated carbon (PAC) was studied under different conditions. The term sorption is used when a combined effect of adsorption and absorption takes place. The encouraging preliminary results were followed by the use of the design of experiments method [7] in order to predict the sorption capacity of the PU-activated carbon system, without going through laborious experimentation.

2. Theory

2.1. Mathematical modeling

2.1.1. The design of experiments method

As a part of the operation of test results, the method of design of experiments seems appropriate for studying the influence of various parameters. Modeling requires the creation of a design matrix. For each input factor, it is necessary to note the levels or the experimental points of the different factors. In our case, the realistic levels were as follows:

1. The time factor that takes the following levels: 5–15–30–60–120–180–2,880 min.
2. For the agitation speed factor, the two levels: 0–200 rpm.
3. For the percent weight of the activated carbon factor, the three levels: (0.00–3.00–5.81)%.
4. For the density factor, several levels.

The design of experiments method requires coding factor levels; the high and low levels can be generically coded as +1 and −1. Input factors may have various levels and can be expressed in different units, it is necessary to eliminate units to obtain dimensionless coded values to enable factor comparison. This coding can be done through the following relationship:

$$x_i = \frac{u_i - \left(\frac{u_{\min_i} + u_{\max_i}}{2}\right)}{\left(\frac{u_{\max_i} - u_{\min_i}}{2}\right)} \quad (1)$$

with u_i: real intermediate value to be coded; $u_{\min_i}$: real value corresponding to the low level; $u_{\max_i}$: real value corresponding to the high level; x_i: coded value of the effect.

An answer depends on the interaction of all factors involved. Analytically, the dependence between the response and the factor can only exist if proportionality exists between them, which can be characterized by factor $a_{(p,i).}$ Hence, it is possible to write the following:

$$Y_{(n,i)} = X_{(n,p)} \cdot a_{(p,i)} \quad (2)$$

with n: the number of experiments; p: the number of factors; i: the number of responses.

The answer $Y_{(n,i)}$ is a function of several factors, which themselves have different levels; it is then necessary to express this in a matrix form (X: effects matrix).

The solution giving the values of the coefficients can be expressed by the following formula from the matrix calculation theory:

$$\text{Coefficients} = (X^t X)^{-1} X^t Y \quad (3)$$

where X^{-1} is inverse matrix, X^t the transpose matrix.

3. Experimental

In this study, experimental tests were performed on the pristine polyurethane foam and on the activated carbon-modified polyurethane foam.

3.1. Preparation of flexible foam

Polyurethanes are remarkable as belonging to the only plastics that can be prepared directly from monomers or prepolymers. They result from the chemical reaction of a polyisocyanate with groups containing mobile hydrogen, primarily of hydroxyl groups also called polyols. The urethane polymerization reaction requires a number of reagents such as polyethers, toluene diisocyanates, blowing agents (methylene chloride, water), catalyst (triethylene diamine, stannous octoate), and surfactants (oil silicone).

The choice of the formulation is very important and depends on the desired final characteristics, such as density (or mass volume), rigidity or flexibility of the final product, porosity, and mechanical properties [8]. The network obtained by the various reactions is generally three-dimensional [9]. For a mixture of all the ingredients with adequate and stoichiometric proportions, processing is carried out as follows:

1. The chemical components are incorporated in a given order and mixed for about five seconds in an agitator with a speed controller and a timer. A good mix, facilitated by the presence of a surfactant, is essential to produce homogeneous foam.
2. Some ten seconds after the start of the operation, a chemical reaction occurs, releasing the inflating gas (CO_2). The gas diffuses into the liquid and gives it a “creamy” consistency.

(3) At the same time of gas production, swelling occurs and viscosity increases. After about two minutes, the maximum expansion occurs. At this stage, in a suitable foam formulation, the residual gas escapes through the top of the block that has gained sufficient strength to maintain its shape.

The strength of the foam continues to increase, and after two and a half to three minutes, the foam completely sets. At this time, the foam takes on its final form. After curing for 24 h and cooling, the final physical and mechanical properties are obtained [10].

For the synthesis of the modified polyurethane foam samples, at first, 10 g of polyol was fully mixed with 0, 0.5, or 1 g of PAC for 15–20 s. Another mixture was prepared with 0.8 g of water, 0.2 g of silicone oil, 0.16 g of triethylenediamine, 0.03 g stannous octoate, and 0.65 g of methylene chloride for 15 s. The two mixtures were mixed for 20–25 s. Then, 7.4 g of TDI (toluene diisocyanate) was added to the new homogenized mixture at an agitation speed of 1,000 rpm for 5 s. Immediately after mixing, the mixer blade was taken out and the final mixture was left for 30 min to provide enough time for the foaming reaction. The whole procedure was performed under ambient temperature (23℃). The mold used was 4.2 cm in diameter and 21 cm in height. Fig. 1 shows the scanning electron microscopy (SEM) image of a 0.0 19 g/cm^3 PU foam synthesized in this way.

The composite foam samples were prepared by varying the weight percent of the PAC added to the initial PU formulation in a bid to increase the sorption capacity of the PU foam-activated carbon system. The PAC was characterized (Table 1) prior to its addition in the composite foam.

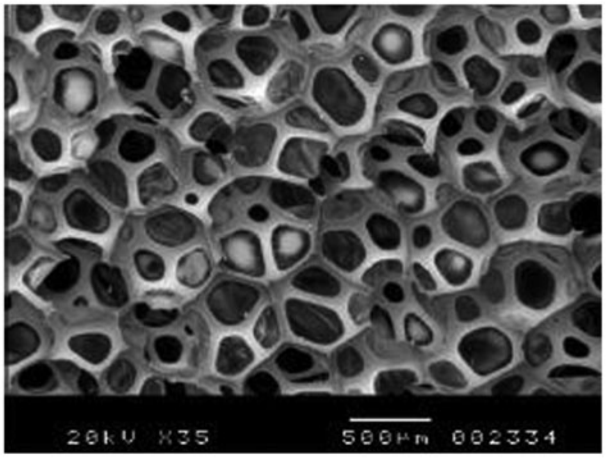

Fig. 1. SEM image of polyurethane foam of density of 0.019 g/cm^3 prepared in the laboratory.

3.2. Study of sorption

The method applied for the measurement of oil sorption capacity of the sorbent is based on the ASTMF726 99 method: Standard Test Method for Sorbent Performance of Adsorbents [11].

For oil sorption tests, 50 mL of crude oil was poured into a 250-mL beaker. The sorbent was weighed and the value recorded, then it was immersed into the oil. After an immersion time *t* (min ± 20 s), the sorbent was removed and allowed to drain for 30 ± 3 s. The saturated sorbent was then immediately transferred to a pre-weighed weighing bottle and weighed. The capacity sorption of oil was calculated as follows:

$$\text{Oil sorption}\left(\frac{\text{g}}{\text{g}}\right) = \frac{m_{\text{sorbed}}}{m_0}\left(\frac{\text{g}}{\text{g}}\right) = \frac{m_t - m_0}{m_0} \quad (4)$$

where m_0 is the initial dry weight of a sorbent, m_t is the weight of sorbent with sorbed oil. All tests were performed at 23℃ in triplicate and the average values of the three runs were used for calculations.

3.3. Oil recovery experiment and sorbent regeneration

For the recovery of the sorbed oil, a compressive force was applied to the sorbent to extract maximum of oil while maintaining intact the physical characteristics of the foam samples. The regeneration of the sorbents was applied using a chemical method used by Nikkhah et al. [12]. The used sorbents were immersed in toluene in a 250-ml beaker and then mixed. After that, sorbent cylinders were taken out and washed by petroleum ether for 3 times. Then, the samples were dried in an oven for 1 h at 65℃.

4. Results and discussion

The results shown in Table 2 allowed to work in an unconventional plan. The graphical representation of the three level “response-surface” design or “iso-responses” was obtained with three data, two effects, and one answer.

After the sorption tests, the sorbed crude oil was recovered according to the method described above. For the foam samples investigated, the maximum recovery rate was 96.7% for the 5.81 wt.% PAC sample, while for the 3 wt.% PAC and the 0 wt.% PAC samples, the rates were 94.6 and 93%, respectively.

The coded values of each factor obtained for the twenty-nine samples studied are shown in Table 3.

Applying the DOE method, the following mathematical model was obtained:

Table 1
Properties of the PAC used in PU formulation

Iodine number (mg/g)	816.05
Methylene blue number (mg/g)	297.3
pH at the point of zero charge	6.45
Specific surface area S_{BET} (m^2/g)	1,031
Surface area available to methylene blue S_{BM} (m^2/g)	800
Density (g/cm^3)	0.33

$$Y = 35.826 + 10.400\,x_1 + 2.461\,x_2 + 1.405\,x_3 + 12.118\,x_4 - 19.588\,x_1^2 + 1.463\,x_1 x_2 + 2.579\,x_1 x_3 + 8.515\,x_1 x_4 + 1.122\,x_2 x_3 - 1.458\,x_2 x_4 - 6.180\,x_3 x_4 \qquad (5)$$

where Y is the sorption capacity; x_1 is the time factor; x_2 is the agitation speed factor; x_3 is the weight percent factor of the PAC; and x_4 is the density factor.

4.1. Evaluation of the quality of the mathematical model

In the present work, the regression analysis was based on the evaluation of two parameters: R^2 (squared correlation coefficient) and Q^2 (cross-validated correlation coefficient) [13]. The quality of the model obtained can be evaluated by static tests represented in Fig. 2.

The R^2 coefficient evaluates the degree of explanation of the mathematical model applied (predicted responses) compared with measured responses. A good model should have an R^2 value close to unity. In this study, R^2 attained 0.998. Q^2 is very similar to R^2; it is sometimes called predictive R^2. It can be negative for very bad models. Values close to unity indicate similarly well-fitting models to experimental data. In this study, Q^2 reached 0.816. The third column in the figure represents the validity of the model. When the validity of the model is larger than 0.25, there is no lack of adjustment of the model, meaning that the

Table 2
Variation of the sorption capacity (sorbed weight per unit weight of sorbent) vs. contact time, agitation speed, PAC weight percent, and density

Experiment number	Time *t* (min)	Agitation speed (rpm)	Activated carbon weight percent PAC wt%	Foam density R_o (g/cm^3)	Sorption capacity m_{sorbed}/m_0 (g/g)
1	5	0	0	0.0171	2.251
2	15	0	0	0.0176	3.094
3	30	0	0	0.0174	3.123
4	60	0	0	0.0147	2.408
5	120	0	0	0.0151	3.455
6	180	0	0	0.0163	5.800
7	2,880	0	0	0.0165	6.989
8	5	0	3	0.015	2.477
9	15	0	3	0.017	3.140
10	60	0	3	0.021	6.371
11	2,880	0	3	0.017	12.498
12	5	0	5.81	0.015	3.461
13	15	0	5.81	0.014	3.371
14	30	0	5.81	0.017	4.250
15	5	200	0	0.019	4.277
16	15	200	0	0.015	2.302
17	30	200	0	0.018	5.294
18	60	200	0	0.014	2.589
19	120	200	0	0.015	6.073
20	2,880	200	0	0.014	7.757
21	5	200	3	0.015	5.273
22	15	200	3	0.017	5.681
23	30	200	3	0.017	8.597
24	60	200	3	0.014	7.204
25	2,880	200	3	0.011	13.447
26	5	200	5.81	0.016	9.111
27	15	200	5.81	0.02	7.723
28	30	200	5.81	0.018	9.213
29	2,880	200	5.81	0.017	30.205

Table 3
Coded values of the different terms of the model

Experiment number	t	[a]Agis	% PAC	R_o	t × Agis	t × % PAC	t × R_o	t × t	Agis × % PAC	Agis × R_o	% PAC × R_o
1	−0.997	−1	−1	−0.432	0.997	0.997	0.431	0.994	1.000	0.432	0.432
2	−0.990	−1	−1	−0.392	0.990	0.990	0.388	0.980	1.000	0.392	0.392
3	−0.979	−1	−1	−0.408	0.979	0.979	0.399	0.958	1.000	0.408	0.408
4	−0.958	−1	−1	−0.624	0.958	0.958	0.598	0.918	1.000	0.624	0.624
5	−0.917	−1	−1	−0.592	0.917	0.917	0.543	0.841	1.000	0.592	0.592
6	−0.875	−1	−1	−0.496	0.875	0.875	0.434	0.766	1.000	0.496	0.496
7	1	−1	−1	−0.480	−1.000	−1.000	−0.480	1.000	1.000	0.480	0.480
8	−0.997	−1	0.0327	−0.600	0.997	−0.033	0.598	0.994	−0.033	0.600	−0.020
9	−0.99	−1	0.0327	−0.440	0.990	−0.032	0.436	0.980	−0.033	0.440	−0.014
10	−0.958	−1	0.0327	−0.120	0.958	−0.031	0.115	0.918	−0.033	0.120	−0.004
11	1	−1	0.0327	−0.440	−1.000	0.033	−0.440	1.000	−0.033	0.440	−0.014
12	−0.997	−1	1	−0.600	0.997	−0.997	0.598	0.994	−1.000	0.600	−0.600
13	−0.99	−1	1	−0.680	0.990	−0.990	0.673	0.980	−1.000	0.680	−0.680
14	−0.979	−1	1	−0.440	0.979	−0.979	0.431	0.958	−1.000	0.440	−0.440
15	−0.997	1	−1	−0.280	−0.997	0.997	0.279	0.994	−1.000	−0.280	0.280
16	−0.99	1	−1	−0.600	−0.990	0.990	0.594	0.980	−1.000	−0.600	0.600
17	−0.979	1	−1	−0.360	−0.979	0.979	0.352	0.958	−1.000	−0.360	0.360
18	−0.958	1	−1	−0.680	−0.958	0.958	0.651	0.918	−1.000	−0.680	0.680
19	−0.917	1	−1	−0.600	−0.917	0.917	0.550	0.841	−1.000	−0.600	0.600
20	1	1	−1	−0.680	1.000	−1.000	−0.680	1.000	−1.000	−0.680	0.680
21	−0.997	1	0.0327	−0.600	−0.997	−0.033	0.598	0.994	0.033	−0.600	−0.020
22	−0.990	1	0.0327	−0.440	−0.990	−0.032	0.436	0.980	0.033	−0.440	−0.014
23	−0.979	1	0.0327	−0.440	−0.979	−0.032	0.431	0.958	0.033	−0.440	−0.014
24	−0.958	1	0.0327	−0.680	−0.958	−0.031	0.651	0.918	0.033	−0.680	−0.022
25	1	1	0.0327	−0.920	1.000	0.033	−0.920	1.000	0.033	−0.920	−0.030
26	−0.997	1	1	−0.520	−0.997	−0.997	0.518	0.994	1.000	−0.520	−0.520
27	−0.990	1	1	−0.200	−0.990	−0.990	0.198	0.980	1.000	−0.200	−0.200
28	−0.979	1	1	−0.360	−0.979	−0.979	0.352	0.958	1.000	−0.360	−0.360
29	1	1	1	−0.440	1.000	1.000	−0.440	1.000	1.000	−0.440	−0.440

[a] Agis stands for agitation speed.

error of the model is in the same order as the pure error. When the validity of the model is less than 0.25, the error of the model is significantly larger than the pure error (reproducibility). A model validity value of 1 represents a perfect model. The validity of the model used attained 0.967.

Reproducibility, or closeness of the agreement between the results of measurements of the same measure, is the variation of the response under the same conditions (pure error), often in the central points, compared with all the variation of the response. A reproducibility value of 1 represents perfect reproducibility. In this case, the reproducibility was 0.975.

4.2. Comparison between experimental and predicted values

Fig. 3 depicts the differences between experimental and predicted values. It can be observed that these differences are not very significant. The model (Eq. (5)) may be assumed to be correct.

4.3. Effect of the investigated parameters on the sorption capacity

Fig. 4 shows the sorption capacity of crude oil (g sorbed per gram foam) as a function of four parameters affecting sorption.

These figures show the significant effect of the four parameters on the amount of oil sorbed per unit weight of sorbent or the sorption capacity (g/g). It can be observed that the increase in one parameter or of all of them together results in an increase in the sorption capacity. It should be noted, however, that the time factor does not behave accordingly beyond the 1,872 min time period (Fig. 4(c)) where a decrease in the sorption capacity was observed. This can be explained by the existence of the term $-x^2$ (x^2 = time^2)

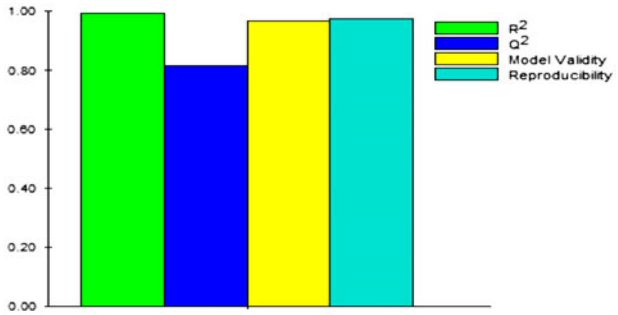

Fig. 2. Representation of the model adjustment coefficients.

in the mathematical model, resulting in an n-shaped parabola. At the 1,872 min contact time, the sorption capacity reached the value of 37.183 g/g for an agitation speed of 100 rpm, a density of 0.023 g/cm^3, and a PAC weight percent of 2.905%. The decrease in the sorption capacity after this time period may be explained by several factors acting individually or jointly, such as the concentration gradient build up between the sorbed phase and the bulk phase, repulsive forces at the surface phase, or partial molecular rearrangement of the complex crude oil mixture molecules from the surface phase to the bulk phase, lowering the free energy of the system [14,15].

4.4. Analysis of the simultaneous effect of the four factors on the response distribution

The experimental area is defined from the variation of the four factors but the graphical illustration of the variation of the response under the action of four factors poses some problems. However, the response surface methodology is recommended to complete this study using two axes for the factors and the third axis for the answer. The response surfaces are three-dimensional graphics, the horizontal plane of the figure shows the range of variation of two factors, while the vertical axis materializes the variation of the response from the model. If more than two factors are considered, it is necessary to maintain the values of the latter at a constant level (intermediate value) where the variations are neither visible nor shown on one of the horizontal axes. The principle is to vary both factors simultaneously while fixing the other two with these intermediate values ($X = 0$; in coded value). The graphical analysis of the model is to illustrate its equation in two forms: response surfaces and iso-response curves [16]. The resulting model consists of first-degree, second-degree terms, and interacting other terms.

For the following Figs. 5–7, figure (a) represents the response in the form of surface and figure (b) the projection of this surface onto a plane.

4.5. Analysis of the sorption response as a simultaneous function of x_2 and x_4

Fig. 5 depicts two different ways of representing the responses. The first one shows the response in space with a surface, while the other shows the projection of this surface on a plane called iso-responses based on agitation speed and density (R_0) using constant values of time (1,440 min) and percent weight of PAC (2.905%). These representations were obtained from the global model of the sorption behavior of the foam.

Horizontal analysis of the iso-responses shows that for different levels of density R_0, the sorption capacity increases slowly for the whole agitation speed range (from 0 to 200 rpm). Agitation speed

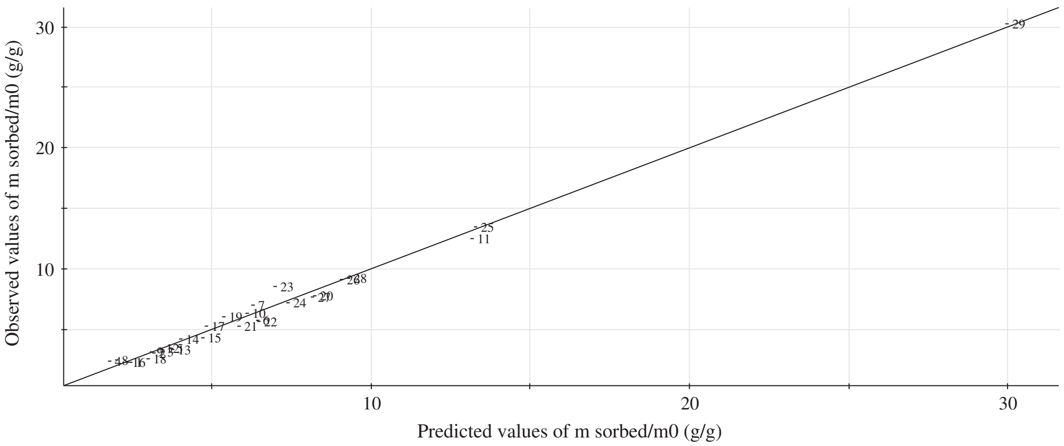

Fig. 3. Differences between observed and predicted values of the sorption capacity of crude oil.

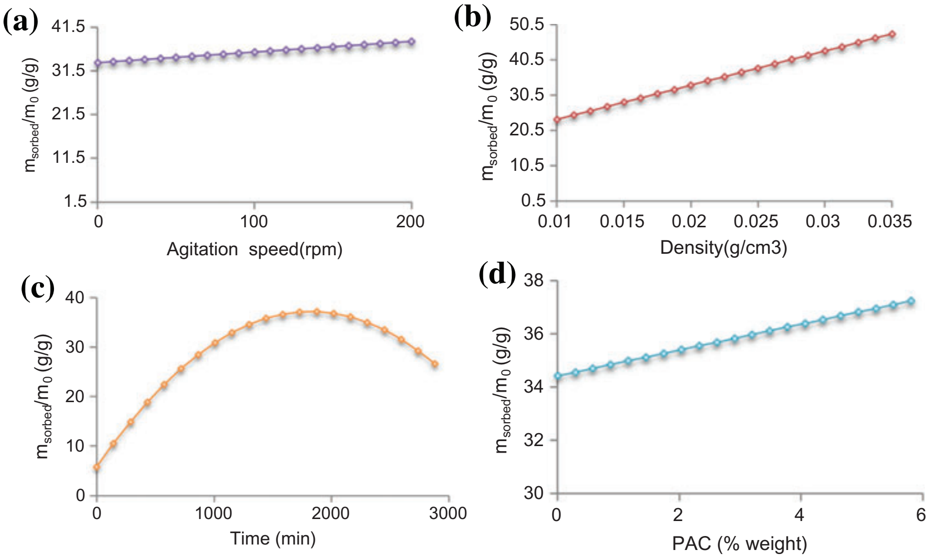

Fig. 4. (a) Effect of agitation speed on sorption capacity of crude oil, (b) Effect of foam density on sorption capacity of crude oil, (c) Effect of time on sorption capacity of crude oil, and (d) Effect of PAC weight percent on sorption capacity of crude oil.

increase does not make much difference; in contrast, the analysis of the vertical iso-responses reveals that for an agitation speed equal to 100 rpm, sorption increases in a considerable way where it goes from 25.6 to 48.8 g of sorbed oil/g of sorbent.

The analysis of the first iso-response provides information on the density parameter acting directly on the sorption capacity of the foam. Simply, an increase in foam density directly affects its sorption behavior. Noting that maximum sorption uptake (48.8 g/g) corresponds to a density equal to 0.035 g/cm^3 and an agitation speed of 200 rpm by fixing the other two factors in the center (PAC wt% = 2.905 and time = 1,440 min).

4.6. Analysis of the sorption response as a simultaneous function of x_2 and x_3

A similar study can be done with the methodology of response surfaces and iso-responses by analyzing the variation of the response as a function of agitation speed and weight percent of PAC (Fig. 6). The parameters kept constant are time (1,440 min) and density (0.023 g/cm^3).

The analysis of the second iso-response reveals the influence of the agitation speed and the PAC percent weight on the foam sorption capacity. In this case, the maximum amount sorbed is equal to 40.26 g/g for an agitation speed equal to 200 rpm and for a PAC percent weight of 5.81%.

4.7. Analysis of the sorption response as a simultaneous function of x_1 and x_3

Similarly to the previous cases, we carried out the third combination between the parameters of time and PAC percent weight. Fig. 7 illustrates the variation of the response as a function of these parameters while keeping the agitation speed and density constant at 100 rpm and 0.023 g/cm^3, respectively.

The two illustrations explain the behavior of the raw foam or the PAC–PU material vs. time. For the optimum time and maximum activated carbon weight percent, one gram sample of foam was observed to sorb 39.6 g of crude oil at an agitation speed set at 100 rpm and a density of 0.023 g/cm^3.

4.8. Analysis of the sorption response as a simultaneous function of x_1 and x_4

For the fourth combination, time and agitation speed were varied, while density and PAC percent weight were fixed at 0.023 g/cm^3 and 2.905%. The response surface is depicted in Fig. 8.

In this study, agitation speed does not play as a significant role as the other factors. Increasing the agitation speed will increase energy expenses as well as time consumption.

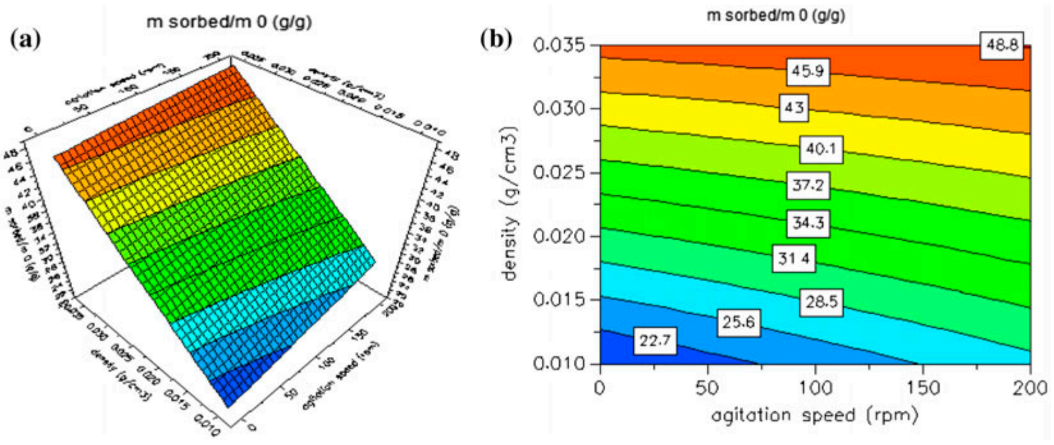

Fig. 5. Sorption response as a simultaneous function of x_2 and x_4. (a) surface response and (b) iso-response.

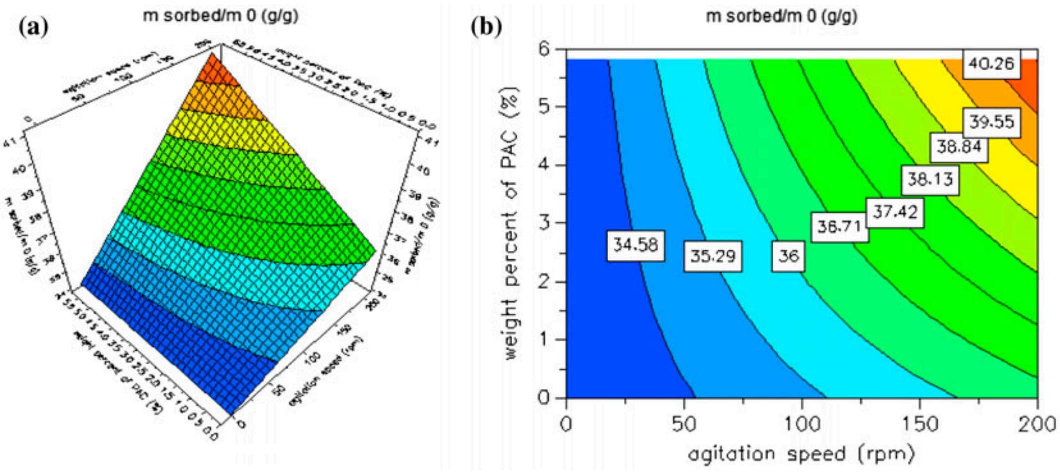

Fig. 6. Response as a simultaneous function of x_2 and x_3. (a) surface response and (b) iso-response.

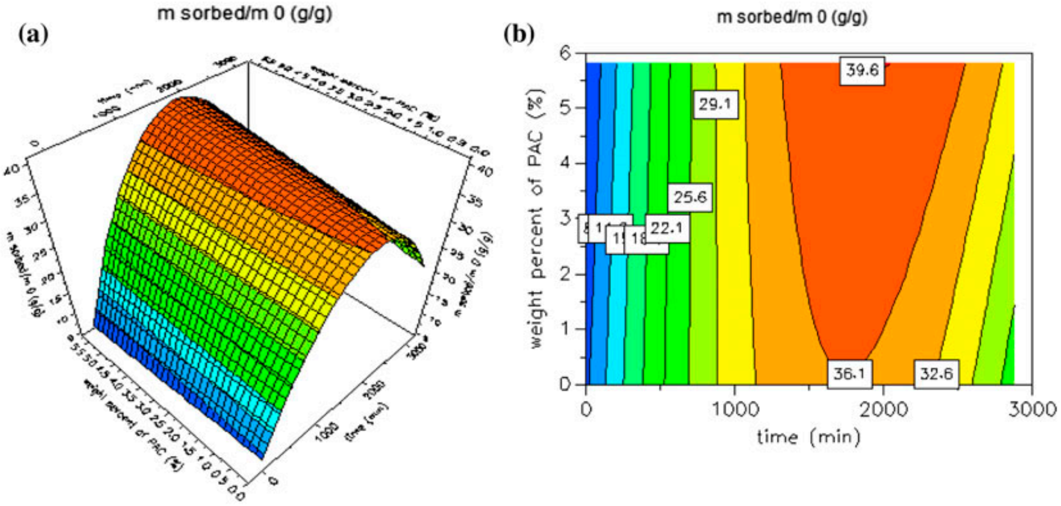

Fig. 7. Sorption response as a simultaneous function of x_1 and x_3. (a) surface response and (b) iso-response.

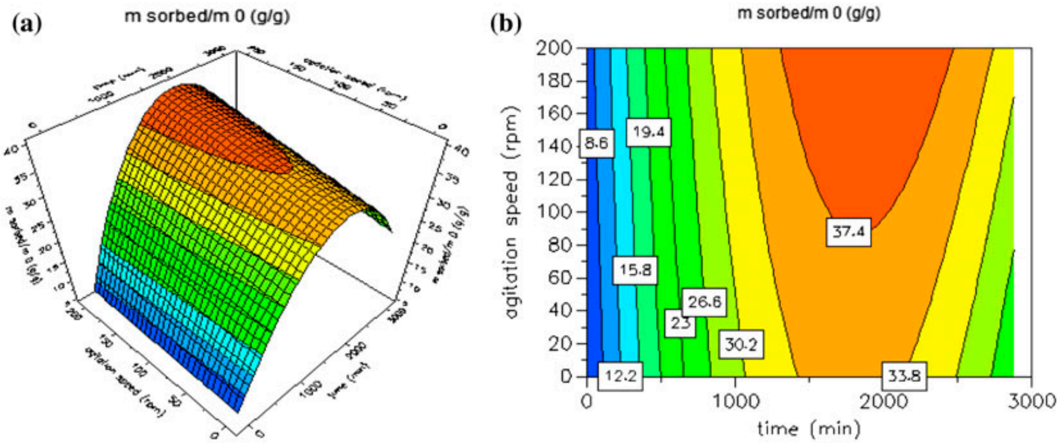

Fig. 8. Response (sorption) as a simultaneous function of x_1 and x_4. (a) surface response and (b) iso-response.

5. Conclusion

Adsorption is a surface phenomenon, which means that extending surface contact between the adsorbent and the adsorbate increases the adsorption capacity. The incorporation of PAC to the initial ingredients of the polyurethane foam in order to prepare a porous cellular form was the aim of this study. A good experimental protocol is provided by the design of experiments method, which has been used to study the above phenomenon to model or predict the response (the results) as a function of variability factors for the proposed model. The validity of the model proposed attained 0.967 while reproducibility reached 0.975. In this study, a crude oil sorption capacity of 50.42 g/g was obtained for the following values of the factors considered: time, agitation speed, PAC percent weight, and density of 1,872 min, 200 rpm, 5.81%, and 0.035 g/cm^3, respectively. Agitation speed was observed to have a small but positive effect on the oil sorption process. This study showed that the polyurethane foam–PAC composite may be used to efficiently sorb crude oil in an economical way.

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