

Application of Box-Behnken Experimental Design in the Optimization of Cleaning Microfiltration Ceramic Filters Using Sodium Hydroxide

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Abstract

Chemical cleaning has been reported as the best method to restore ceramic filters flow rate by removing fouling agents. Even though there are several chemicals that can be used as cleaning agents, sodium hydroxide (NaOH) has been widely used as a cleaning agent. Literature reports that main factors of this cleaning are sodium hydroxide concentration, treatment temperature and contact time. However, the most significant factors have not been determined nor the interactions between them. The aim of this study was to determine the most significant parameter and the interactions between sodium hydroxide concentration, treatment temperature and contact time. This was done using Box-Behnken experimental design. The results, based on ANOVA analyses, showed that temperature is the most significant factor and that interaction between sodium hydroxide concentration and treatment temperature is the most significant interaction.

Keywords

Chemical Cleaning, Ceramic Filters, Microfiltration, Box-Behnken, Optimization

1. Introduction

Fouling is the main drawback of ceramic filters in water treatment and is materialized by the diminishing of filters flow rate. Therefore, researchers have identified various processes to restore filter's initial flow rate by removing fouling agents. These processes are included in what is called cleaning processes. Cleaning

ceramic filters is done by applying a force and based on the nature of the applied force, cleaning can be either physical or chemical [1]. Hydraulic and mechanical forces are related to physical cleaning [2], while dispersion, hydrolysis, peptization, saponification, solubilization, and chelation are related to chemical cleaning [3]. Physical cleaning includes flushing/rinsing, backwashing/backflushing, backpulsing, air enhanced backflushing, ultrasound and electric field [4] [5]. While chemical cleaning is characterized by the usage of chemical agents; there are several categories of cleaning agents such as caustics (e.g., sodium hydroxide), alkalis (e.g., carbonates, hydroxides), acids (e.g., sulfuric acid, oxalic acid), metal chelating agents (e.g., ethylenediaminetetraacetic acid), surfactants (e.g., alkyl sulfate, sodium dodecyl sulfate), enzymes (e.g., proteases and lipases), disinfectants (e.g., peroxyacetic acid, hydrogen peroxide, chlorine) [3] [6]. Nevertheless, alkaline solutions, particularly sodium hydroxide one, have been widely used with best efficiency [4]. Even though physical cleaning is faster than chemical cleaning, it is less efficient particularly when the fouling is irreversible [7] [8]. Therefore, many researchers have been focused on this later cleaning. Their results showed that factors that influence chemical cleaning are chemical agent concentration, treatment temperature and contact time. However, results reported in the literature showed contradictory conclusions on the influence of these parameters on the efficiency of chemical cleaning. Indeed, some have shown that increasing chemical concentration decreases cleaning efficiency [3]. While others have shown the opposite [3]. This controversy is the result of the fact that these researches have been carried out without evaluating the interactions between parameters. It is therefore essential to apply an experimental design. Among all existing experimental designs, Box-Behnken Design (BBD) offers several advantages compared to other designs. BBD has the advantage that it provides information exclusively on the effect of experiment variables or factors and overall experimental error in a minimum number of required runs [9] [10]. It is a three-level second-order spherical design with all points lying on a sphere, providing more flexibility in choosing designs for a specified number of factors [11]. Based on this, Box-Behnken experimental design has been applied to determine the optimum of sodium hydroxide cleaning for microfiltration ceramic filters. In addition, most significant factors of sodium hydroxide cleaning have been determined as well as all the interactions between different factors.

2. Materials and Methods

2.1. Fouled Ceramic Filters

In this study, 20 mL fouled microfiltration cylindrical ceramic filters were used. These fouled filters were obtained after a filtration test involving the retention of colloidal materials. The Filtration test consists of emptying the fulfilled filter and noting the volume of treated water obtained and the time required to empty them. The flow rate was then obtained by dividing the volume obtained by the time necessary to empty the filter. The flow rate of the first trial (first complete empty) is

considered as the filter initial flow rate F1. Filtration test ended when the initial filter's flow rate had decreased by 80%. A number has been assigned to each filter.

2.2. Box-Behnken Experimental Design

2.2.1. Matrix Design

Matrix design was made up using Statgraphics 16 centurion software and was based on a three-factor and three-level design generating thus 15 experiments (12 edges and 3 centre points). The centre points are utilized to evaluate the experimental error and the reproducibility of the data and are experiments where all the factors are in their intermediate level. Level of each factor was as follow: 0.30 mol/L, 0.65 mol/L and 1 mol/L; 5 min, 12.5 min and 20 min and 25 °C, 50 °C and 75 °C respectively for concentration, time and temperature. These values correspond respectively to the minimum, intermediate and maximum and were chosen after a screening experiment. **Table 1** shows matrix design.

Table 1. Matrix design.

Experiment number	Concentration (mol/L)	Temps (min)	Température (°C)
1	0.30	12.50	25.00
2	0.65	12.50	50.00
3	0.65	5.00	75.00
4	1.00	20.00	50.00
5	0.65	5.00	25.00
6	1.00	5.00	50.00
7	0.30	5.00	50.00
8	1.00	12.50	75.00
9	0.65	12.50	50.00
10	0.30	20.00	50.00
11	0.65	20.00	25.00
12	0.65	20.00	75.00
13	0.30	12.50	75.00
14	1.00	12.50	25.00
15	0.65	12.50	50.00

2.2.2. Experimental Setup

Using sodium hydroxide pellets, sodium hydroxide solutions were prepared at different concentrations related to the matrix design. For each experiment, 250 mL of each solution was poured into a 500 mL beaker in which was immersed a 20 mL fouled cylindrical ceramic filter and a magnet bar. Then, the beaker and its

content were placed on a heating magnetic stirrer that was previously calibrated to the desired temperature. With a stopwatch, the contact time was controlled; each experiment was carried out in triplicate. After cleaning, the beaker and its content were allowed to cool at ambient temperature. Afterward the filter was removed from the beaker and rinsed with distilled water and kept at ambient temperature.

2.2.3. Optimization

The experimental sequence was randomized in order to minimize the effects of uncontrolled factors. The outcome of each experimental run was analysed with ANOVA and the response was correlated with three input factors for the flow rate recovery of microfiltration ceramic filters through an empirical second-degree polynomial equation as given by the following equation:

$$Y_i = \beta_0 + \sum \beta_i x_i + \sum \beta_{ii} x_{ii} + \sum \beta_{ij} x_i x_j + \varepsilon \quad (1)$$

where, Y_i is the predicted response (flow rate recovery), β_0 the constant coefficient, β_i the linear coefficients, β_{ij} the interaction coefficients and β_{ii} the quadratic coefficient. ANOVA was used to optimize the system by estimating the statistical parameters.

The polynomial model (Equation (1)) is validated by comparing, on the one hand, the theoretical and experimental values of the response (determination coefficient (R^2)). And on the other hand, by evaluating the Absolute Average Deviation (AAD) which must respectively be greater than or equal to 0.8 and between 0.0 and 0.3 [12]-[14].

2.3. Filtration Test

The cleaned filters were therefore immersed in a 500 mL beaker containing distilled water for 24 h. Afterward, they were placed up in a funnel that was placed in a test tube. The filters were filled with water containing colloidal materials that was used to foul the filters. They were allowed to filter until they were emptied. Each time the filter emptied, the time required and the volume of filtrate were recorded. The flow rate was then obtained by dividing the volume obtained by the time necessary to empty the filter. Moreover, the flow rate was noted as F2. Cleaning efficiency was obtained by applying the following formula:

$$P = \left(100 - \frac{F1 - F2}{F1} \right) \times 100 \quad (2)$$

where, P is the flow rate recovery, F1 filter initial flow rate and F2 cleaned filter flow rate.

3. Results and Discussion

3.1. Matrix Design Results

Table 2 shows the experimental (Y1) and the theoretical (Y2) values of flow rate recovery. The results obtained in this table permitted us to calculate determination

coefficient R^2 that was equal to 0.84 and AAD that was equal to 0.05. These show that the polynomial model is validated.

Table 2. Matrix design results.

N°	Concentration (mol/L)	Temps (min)	Temperature (°C)	Y1 (%)	Y2 (%)
1	0.30	12.50	25.00	37.69	35.97
2	0.65	12.50	50.00	13.06	15.47
3	0.65	5.00	75.00	56.08	47.15
4	1.00	20.00	50.00	20.38	21.46
5	0.65	5.00	25.00	17.39	20.18
6	1.00	5.00	50.00	12.27	19.48
7	0.30	5.00	50.00	35.00	33.92
8	1.00	12.50	75.00	49.63	51.35
9	0.65	12.50	50.00	13.06	15.47
10	0.30	20.00	50.00	33.16	25.94
11	0.65	20.00	25.00	10.38	19.31
12	0.65	20.00	75.00	44.81	42.02
13	0.30	12.50	75.00	24.31	34.32
14	1.00	12.50	25.00	10.03	0.02
15	0.65	12.50	50.00	20.28	1.47

3.2. Optimization

3.2.1. Variance Analysis

Table 3 shows variance analysis obtained using ANOVA. It shows that only temperature has a significant effect at the 95% confidence level on the cleaning. Indeed, temperature is the lone factor that has a p-value less than 0.05. According to the above result, temperature would therefore be the only factor that would positively influence microfiltration ceramic filters cleaning.

Table 3. ANOVA variance analysis results.

Source	Sum of squares	Df	Squares mean	F-Ratio	p-value
Concentration	179.08	1	179.08	1.47	0.28
Time	18.03	1	18.03	0.15	0.72
Temperature	1233.55	1	1233.55	10.10	0.02

3.2.2. Factors Contribution

With the polynomial model validated factors contribution can therefore be

evaluated. The results are shown in **Figure 1**, which represents the Pareto diagram. This diagram shows that temperature has a positive effect, while concentration and time have a negative effect. It is also shown that negative effect of concentration is higher than the one of time. The positive effect of temperature suggests that when increasing temperature cleaning efficiency increases. Moreover, the negative effect of the two other factors means the opposite. The positive effect of temperature might be explained by the fact that higher temperature favours fouling agent's dispersion [15] [16] changing fouling agents to their higher dispersive form. Indeed, higher temperature will allow the breaking of bonds between the clogging agents and walls of filter's pores.

Concerning factors quadratic effect, **Figure 1** shows that quadratic effect of temperature and its linear one have both positive effects. While the opposite is observed for concentration and time. This means that there exists a range of values where higher concentration and higher time increase cleaning efficiency. Concerning interactions effect, **Figure 1** shows that interaction between concentration and temperature has the most significant effect. Moreover, only interaction between time and concentration has a negative effect. This can be explained by the fact that both of them have a negative effect on cleaning efficiency. Furthermore, it can be concluded that temperature is the factor that have the significant effect on cleaning efficiency.

The model equation obtained after ANOVA analysis was as follow:

$$Y = 107.353 - 1.752A - 0.056B + 115.742C + 0.013A^2 + 1.514B^2 + 1.925C^2 + 0.948AB + 11.302AC - 0.006BC \quad (3)$$

where A = concentration, B = time and C = temperature.

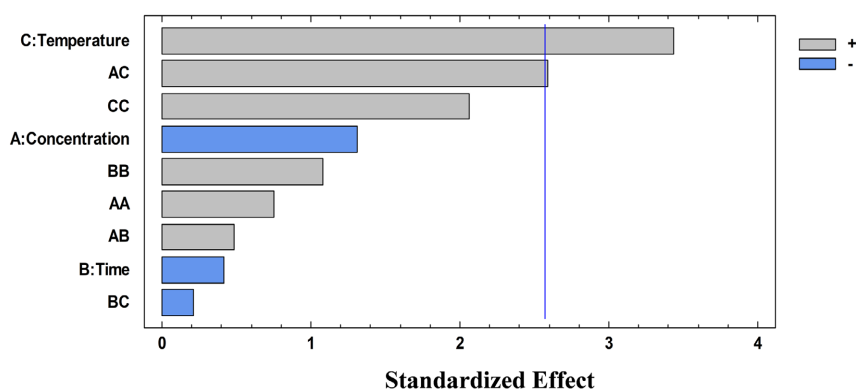


Figure 1. Pareto diagram.

3.2.3. Optimum

Evaluation of factors contribution has shown that temperature is the most significant factor of sodium hydroxide cleaning. Therefore, to determine the optimum it has to be fixed while varying time and concentration. This was done and the results are plotted in **Figure 2** showing the response surface. Temperature was fixed to 70°C.

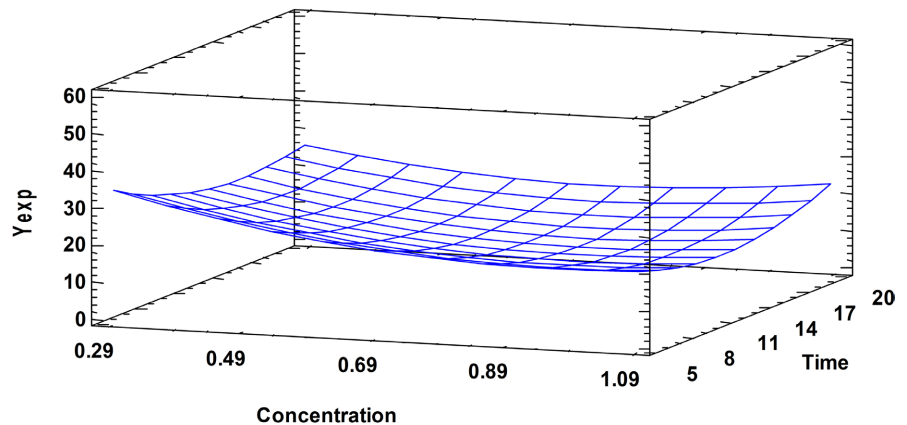


Figure 2. Response surface.

Therefore, optimum flow rate recovery was obtained and its value was 51%. This optimum was achieved with these conditions: concentration 1 mol/L, time 12 min and temperature 75°C. This optimum is in the range (50% - 65%) sodium hydroxide cleaning ceramic filters [3].

4. Conclusion

This work showed the application of the Box-Behnken experiment to optimize sodium hydroxide cleaning microfiltration ceramic filters. The results have shown that temperature is the most significant factor and that interaction between concentration and temperature is the most significant interaction. Information missing in the literature before. Optimum filters flow rate recovery was 51% and is in the range of other similar studies.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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