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Radioactive wastes Conditioning. Optimization of operating parameters by experience plan method

Bouchra El Hilal* and Ahmed El Harfi

Laboratory of Organic & Macromolecular Chemistry, Department of Chemistry, Faculty of Science, University Ibn Tofail, BP 133, 14000, Kenitra, Morocco.

Abstract: The conditioning of exhausted Resins Exchanging Ions (REI) (radioactive wastes generated by nuclear operations) was optimized using a full factorial experiments plan 2^4. Sixteen experiments with a suitable choice of four interpretable variables led to a mathematical model in the form of a first degree polynomial. After analysing the effects, this model showed that the most influential factor on the response (compression strength) is the water/cement ratio (W/C) with a positive effect of (+2.17), the second factor in order is the mixing time with a positive effect of (+1.54). The interaction between the (W/C) and the number of vibration and interaction between the (W/C) and the mixing time also have effects on the response.

Keywords: Radioactive waste, packaging radioactive waste, compressive strength, experiments plan, Nemrodw software.

Introduction

The exploitation of radioactivity in different areas of life becomes more and more intense in contemporary society: Electricity production, radiotherapy, non-destructive control...

Nevertheless, these activities generate radioactive waste which has great hazards to human and its environment.

Among these waste we find ion-exchange resins which are polymers [1] used in water purification circuits of nuclear reactor [2, 3]. After a certain time of use in nuclear reactor, exhausted resins are changed, and they are considered as radioactive waste requiring appropriate management.

The Management of this type of radioactive waste consists of minimizing their harmful effects [4, 5, 6, 7] through their immobilization in solid matrix generally based on cement. The faced challenge, in the immobilization processes, is the construction of matrix that will keep sustainably good mechanical properties (compressive strength) and chemical properties (leaching) [8-16].

In this study we sought to optimize the experimental conditions of conditioning of exhausted ion-exchange resins (REI) - radioactive waste generated by reactor TRIGA MARK II of CNESTEN (Morocco) [17-21] using experience plan method [22-25], based on the following parameters: the water / cement ratio "E / C", the % superplasticizer (addition) "% sup", the number of vibration of the containment test "number of vibration" (vibration performed by a vibrator Type Trakita VR 250D to eliminate air pockets containing in the containment test) and finally the mixing time.

Experimental Section

This study was performed using the Nemrodw software that allows the development of plans of experiments and analysis of experimental results by analysing statistical indicators and specific tools [26] to achieve our objective.

Presentation of the problem

Objective of the study:

Find the best conditions for REI conditioning.

Study Description:

We wish to study the effects of four variables: the percentage of super-plasticizer "adding" (X1), the water/cement ratio "E / C" (X2), the number of the specimen vibration of (X3) and the mixing time (X4) on the compressive strength of the REI containment matrix.

Reply

The measured answer Y is the compressive strength of the REI conditioning matrix after 28 days of confinement.

**Corresponding author: Bouchra ELHILAL*
E-mail address: bouchra-elhilal@hotmail.fr
DOI: http://dx.doi.org/10.13171/mjc.5.1/01603031520/elhilal

Principle

To perform a series of tests arranged in advance so as to determine a minimum of trial and a maximum of precision, the multiple parameters influence on the measured values from the response Y (compressive strength)

Mathematical modeling

The answers are described by a polynomial model [14] of the following form:

Y = b0 + b1 * X1 * X2 + b2 + b3 + b4 * X3 * X4 + b12 * (X1 * X2) + b13 * (X1 * X3) + b23 * (X2 * X3) + b14 * (X1 * X4) * b24 + (X2 * X4) + b34 * (X3 * X4)

With:

- Y: Response to model (compressive strength)
- X1, X2, X3 and X4: studied factors
- X1X2, X1X3, X1X4, X2X3, X2X4, X3X4:

Interactions between factors

- b0, b1, b2, b3, b4, b12, b13, b23, b14, b24, b34: Effects

Experimental Section

The factors evaluated in this study and the levels of studies are presented in the following table

Table 1. Experimental Area.

Factors	X1(% superplasticizer)	X2 (E/C)	X3(Number of vibration)	X4(mixing time)
Unit	-	-	-	second
Level (-1)	0.05	0.1	20	100
Level (+1)	0.4	0.4	40	300

Experimentation plan

The experimental plan is schematically shown in the following table 2:

Table 2. Experimental plan.

N° Exp	Rand	% super-plasticizer	E/C	Number of vibration	mixing time (s)	Compressive strength (MPa)
1	12	0.05	0.10	20	100	9.10
2	7	0.40	0.10	20	100	9.80
3	3	0.05	0.40	20	100	19.20
4	14	0.40	0.40	20	100	19.70
5	15	0.05	0.10	40	100	12.30
6	5	0.40	0.10	40	100	13.20
7	6	0.05	0.40	40	100	15.40
8	16	0.40	0.40	40	100	15.90
9	4	0.05	0.10	20	300	14.50
10	13	0.40	0.10	20	300	14.30
11	**10**	**0.05**	**0.40**	**20**	**300**	**20.00**
12	1	0.40	0.40	20	300	18.50
13	11	0.05	0.10	40	300	18.20
14	8	0.40	0.10	40	300	16.30
15	9	0.05	0.40	40	300	16.70
16	2	0.40	0.40	40	300	17.00

The experimental plan is the translation of a matrix of experiences in natural variable [27]. It is a table containing data directly usable by experimenters.
Table 3, like the matrix of experiments, consisting of N rows and K columns, each of its elements corresponds to the level that the natural variable takes in the experiment.

Results and Discussion

Statistical Analysis of results

To judge the quality of the obtained results, we use the statistical tests because the equation of empirical model alone has only an approximation of reality [28]

Table 3. The descriptive quality of the model.

Standard Error of the response	0.712
R2	0.985
R2A	0.954
R2 pred	0.841
Press	25.939
Number of degrees of freedom	5

The presented results in Table 3 allowed us to analyze the quality and validity of our model, and after we have concluded that all the answers have satisfactory descriptive quality because the correlation coefficient R2 and the adjusted coefficient of determination R2A have the closest values to 1 [29].

Estimation and statistics of coefficients

Table 4. Analysis of the coefficients.

Name	Coefficient	F.Inflation	Standard Deviation	t.exp.	Signif. %
b0	15.631		0.178	87.84	***
b1	-0.044	1.00	0.178	-0.25	80.9%
b2	2.169	1.00	0.178	12.19	***
b3	-0.006	1.00	0.178	-0.04	97.2%
b4	1.306	1.00	0.178	7.34	**
b12	0.019	1.00	0.178	0.11	91.7%
b13	0.019	1.00	0.178	0.11	91.7%
b23	-1.544	1.00	0.178	-8.68	***
b14	-0.369	1.00	0.178	-2.07	9.2%
b24	-1.056	1.00	0.178	-5.94	**
b34	0.119	1.00	0.178	0.67	53.8%

The inflation factor is an important parameter for assessing the quality of the matrix of experiments [30]. From the results shown in Table 4, we find that this factor has a value of 1.00 for all coefficients which allows us to say that the matrix is of a good quality (1.00 ≤ fi≤ 4.00).

Weight graphics:

The influence of factors and their interactions on the response are evaluated through the coefficients of the model [31]. The Pareto chart is an aid tool of interpretation 35. For this purpose, the compressive strength results are shown in the diagram (Figure 1).

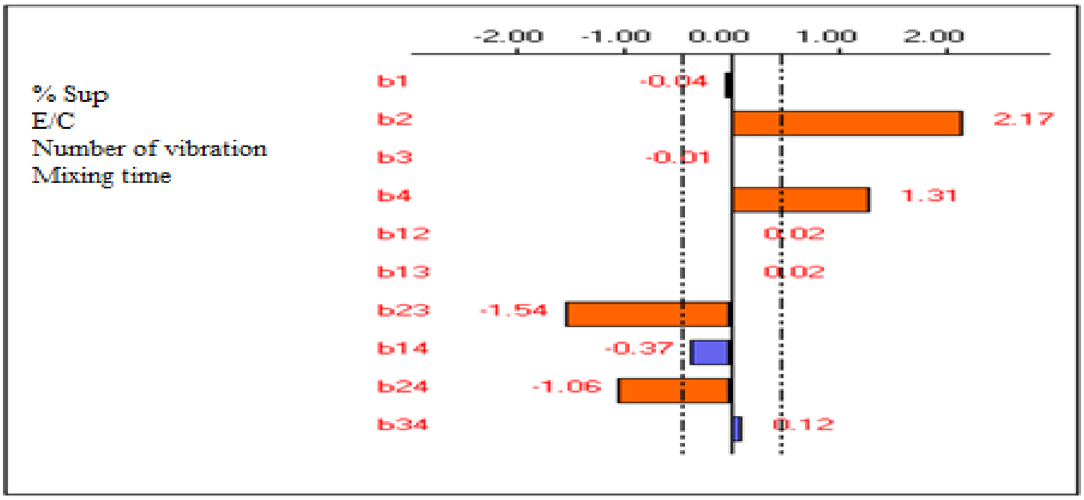

Figure 1. Pareto Chart

From Figure 1, we note that the factors E / C b2 and the mixing time b4 and the interaction between (E / C) / number of vibration b23 and (E / C) / the mixing time b24 strongly acts on the response (compressive strength). By contrast, the interaction between % Super-plasticizer / mixing time b24 has a less pronounced influence.

Graphical study of the effects of the response Y

Method of the right of HENRY

If we represent the function of the effects of the distribution (bj or | bj |) on Gausso-arithmetic papers, we get two graphs: the **Normal Plot** graph with bj and the **Half Normal Polt** graph with | bj |.

Normal Half Normal Plot Plot

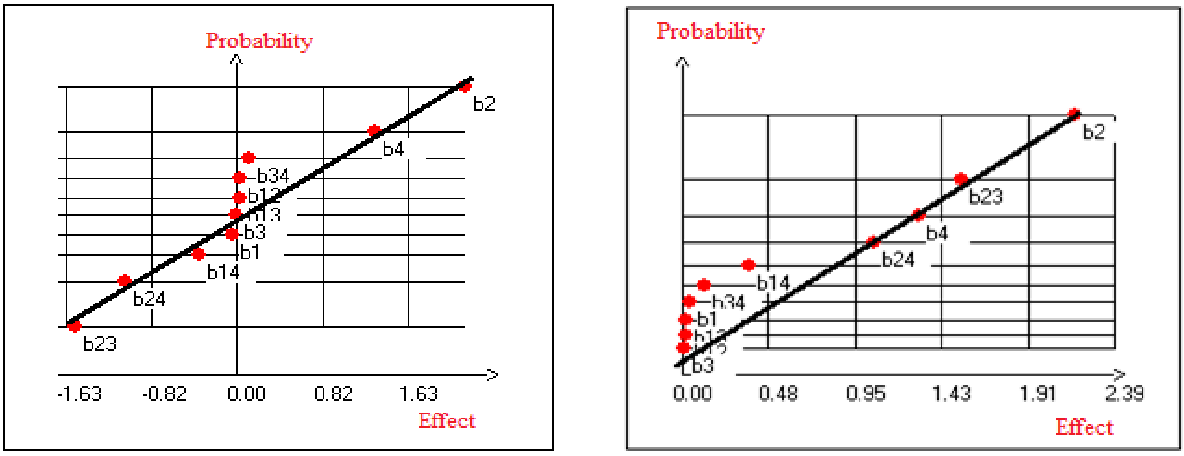

Figure 2: Right of HENRY

The effects that deviate from the right on the Normal Plot below or above (depending on whether the effects are positive or negative) are considered probably active.
The graph of the Half Normal Plot allows to distinguish more clearly the active coefficients but it does not give any sense because we consider here the absolute value of coefficients.
The coefficients b2 and b4, that are respectively the effects of E / C and the mixing time, are probably active, which is consistent with the results obtained in the Pareto chart.

Thus the interaction coefficients b23, b14 and b24 are respectively the interaction (E / C) / Number of vibration, (E / C) / mixing time and % Super-plasticizer / mixing time are probably active.

Study of graphic interactions

A study of the effects is necessary to know the main effects of the factors and the interaction effects between them. An interaction which is a combination of factors does not act independently.

- ***Interaction (E / C) / Number of vibration***

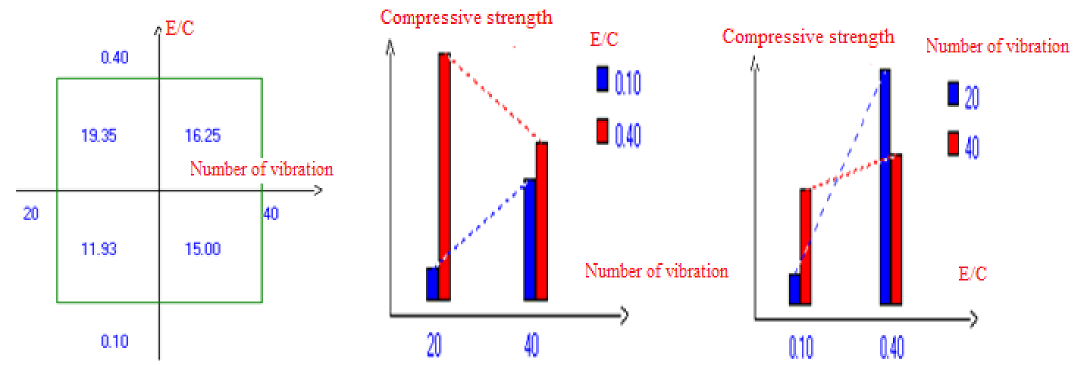

Figure 3. Study of the interaction graph (E / C) / Number of vibration

From Figure 3, we notice that the W / C ratio and the number of vibration significantly affect the response (compression strength) through the interaction between them. To have a resistance to the maximum compression of the confinement matrix, it would be better to use a W / C ratio = 0.4 and to vibrate the specimen 20 times.

- ***Interaction (E / C) / mixing time***

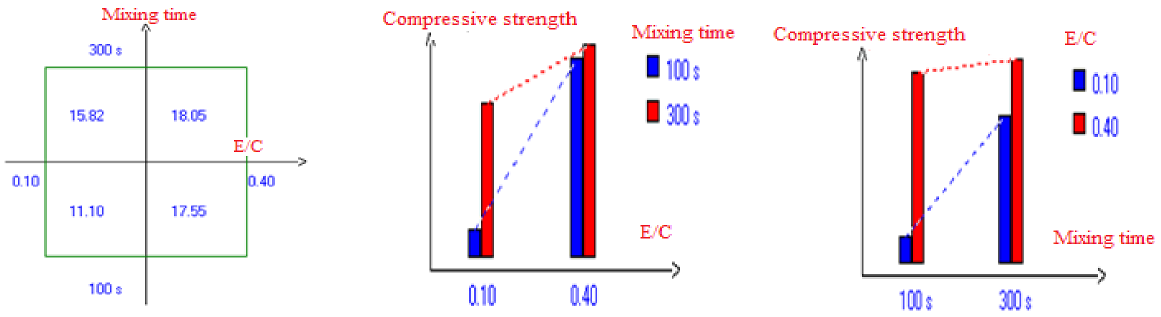

Figure 4. Graphic Study of the interaction (E / C) / mixing time

From the graphs shown in Figure 4, we found that the compressive strength of the radioactive waste containment matrix is influenced by the interaction between the factor W / C and the mixing time factor. Resistance to the maximum compression is recorded for 300 seconds of the mixture and for a W / C ratio equal to 0.4.

- ***Interaction mixing time /% superplasticize***

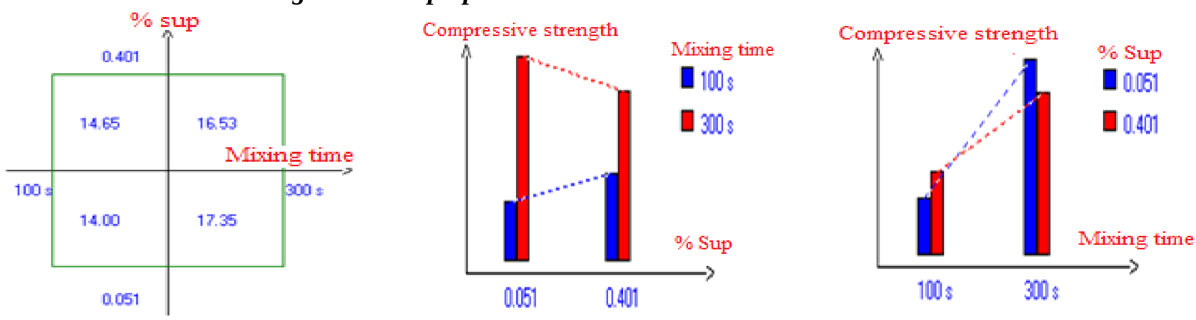

Figure 5. Graphic study of the interaction mixing time /% superplasticizer

According to graphs in Figure 5, the interaction of mixing time / % superplasticizer has an influence on the compressive strength, as long as it is optimal at a rate of 0.05 of superplasticizer and at 300 seconds of mixing.

- ***Interaction% Superplasticizer / (E / C)***

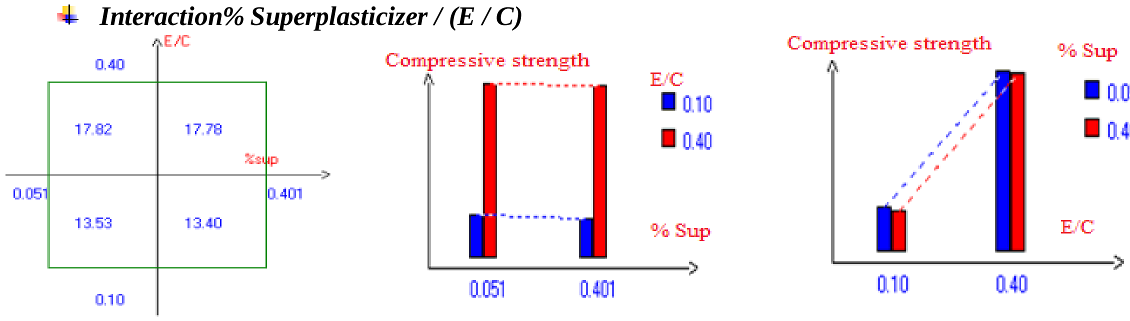

Figure 6. Graphic Study of the interaction % Superplasticizer / (E / C)

- ***Interaction % Superplasticizer / Number of vibration***

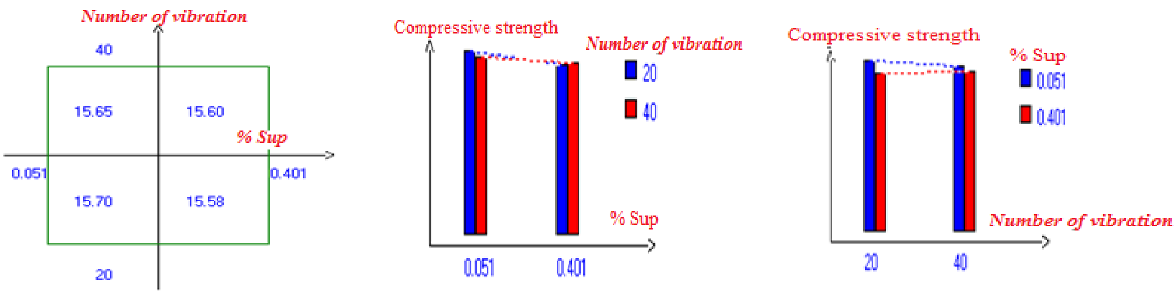

Figure 7. Graphic Study of the interaction% Superplasticizer / Number of vibration

Interaction Number of vibration / mixing time

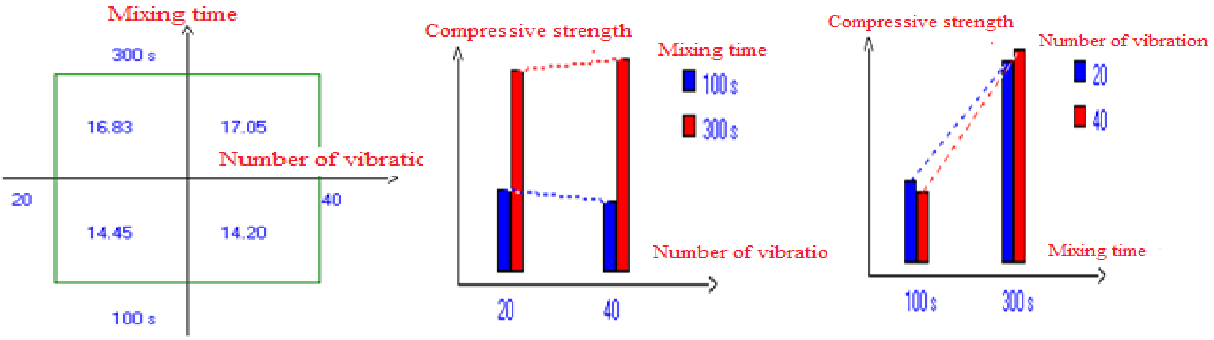

Figure 8. Graphic Study of the interaction, Number of vibration / mixing time

According to figures 6, 7 and 8, we noticed that there is no interaction between the factors and the superplasticizer % (W / C), between the superplasticizer% and the number of vibration and also between the number of vibration and mixing time. This proves that each component has an effect on the resistance to its own compression without taking into consideration the other.

Theoretical correlation of experimental results

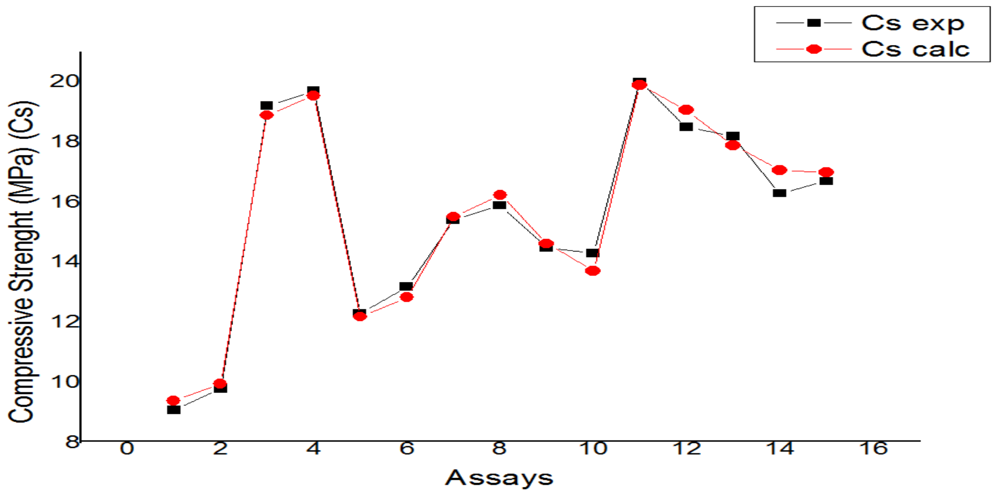

Figure 9. Theoretical correlation of experimental results

Figure 9 illustrates the theoretical correlation of the experimental results and the modeled results, and from this figure we see that the theoretical results coincide with the experimental results. This leads us to say that the chosen model is adequate for this study.

Validation of the plan

The answer Y according to mathematical modeling is:

Y = 0.044 + 2.169X2 15.631- X1 - X3 0006 + 1306 + X4 + 0.019X1X2 0.019X1X3 - X2X3-0.369 X1X4- 1.544 1.056 + X2X4 0.119X3X4

Following the various results presented above, we can say that the optimum formulation, to obtain a maximum of compressive strength, is that of Experiment N° 11 shown in Table 3.

Conclusion

In conclusion of this work undertaken to optimize the experimental conditions of confinement resins exchange ions used in the reactor TRIGA MARK II (CNESTEN-Morocco) (Center national d'Etudes, des Sciences et des Techniques Nucléaire), we have demonstrated that the most influential factors on the response (compressive strength) are the “W/C”, the “mixing time”, the interaction between W/C and number of vibration and finally the interaction between W/C and mixing time.

The experimental conditions as defined by this methodology provided satisfactory results for the optimization of REI immobilization conditions and

**Corresponding author: Bouchra ELHILAL*
E-mail address: bouchra-elhilal@hotmail.fr
DOI: http://dx.doi.org/10.13171/mjc.5.1/01603031520/elhilal

subsequently improved the mechanical performances of the confinement matrix.

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