

Experimental study and modeling the rupture model of a new hydraulic binder based on the combination of inorganic additions by the response surface methodology

M. H.R. Khudhair*^{1,2}, M. S. Elyoubi³, A. Elharfi¹

¹Laboratory of Agro resources Polymers and Process engineering (LAPPE), Team of Macromolecular & Organic Chemistry, Faculty of sciences, Ibn Tofail University, BP 133, 14000, Kenitra, Morocco

²Laboratory of Cement and Quality Control of Amran Cement Plant of Yemen

³Laboratory of Chemistry of Solid State, Faculty of Science, Ibn Tofail University, Kenitra, Morocco

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M. H.R. KHUDHAIR

khudhair.mohammed65@gmail.com
+212 6 33431912

Abstract

The aim of this work is to determine by the experimental design of response surface methodology a model which predicts the compressive strength as a function of the proportion of the constituents of a new hydraulic binder based on a combination of different percentages of Natural pozzolana (PN) and pure limestone (P,Lime). A second-degree polynomial model was therefore used to model the influence of three key parameters which are the water content, the proportion of natural pozzolan and the proportion of pure limestone, related to the compressive strength at 28 days, which represents the model of rupture, while the other components and the operating condition were kept fixed. Our study has three essential phases: in the first phase, the objective is to determine the main physical, chemical, and mechanical characteristics of the materials used, the choice of the method of formulation and the realization of the different mixes. The second phase, which aims to analyze the effects and interactions between the various factors expected, to predict the most influential factor in response on one hand and to highlight it on the correlations and their interactions on the other hand. As for the last phase, it consists of the search for the optimum which is done by the response surface methodology of experimental design of each factor studied and the validation of the model retained.

1. Introduction

For the last 20 years, the statistics have been developed in various fields, including the design of the experiment [1 - 4]. The analysis of data by the design of the experiments allowed us to analyze a large unstructured data sets when the parameters cannot be sufficiently controlled. These analytical methods allowed us to interpret tests already carried out and describe the influences of the parameters involved in a qualitative way [5 - 7]. In order to improve the physical properties of fresh cement paste, such as the water content, the fluidity, the setting time and the compressive strength in the hardened state, we were appalled to the approach of response surface methodology of experimental design, which translates into an iso-response curve. This allowed us to present the variation response in the adjusted space [8 - 12]. The key points of this study, were a strategic component type proportions assuming that the response which is the compressive strength depends only on the variation of three factors/constituents which are the mass fractions of the natural pozzolan (PN) and the pure limestone (P,Lime) which we partially substituted the clinker by the combination of these two additions at different percentages ranging from 5% to 40% by weight of cement with an interval of 5% as well as the report water/cement "W/C" which varies from 0.54 to 0.4.

After the choice of the constituent of the formulation matrix we mixed these constituents and each time we characterized the final product formulated (compressive strength). This approach can be infinite and more costly [13 - 16]. The experimental design methodology allowed us to structure our research, to measure and validate our own hypotheses in order to better understand the phenomena studied [17 - 19]. The use of an experimental design is interesting in that it reduces the number of tests while varying several parameters (factors) at a time, which will allow us to evaluate their influences and their interactions on the response studied. The response surface methodology of experimental design makes it possible to solve optimization problems [20 - 22]. They allow for a polynomial model of the second degree, determine what values to the three already selected factors should be adjusted to achieve the desired response [23 - 25].

The response studied is the compressive strength at 28 days, which presents the rupture model, carried out on prismatic specimens of dimensions (4×4×16) cm³, preserved in water for 28 days. These specimens are broken

in compression, in fact, the mean of different forces of stress of the ruptures. This model of rupture will be noted in MPa. Through the iso-response curves and through the trace of the response surface recalled firstly the effects of main components and the other effects of their interactions, we can determine the operating conditions (values the three factors) which allowed us to formulate a new sustainable hydraulic binder based on the different percentages of natural Pozzolan and pure limestone.

2. Materials and methods

2.1. Materials

2.1.1. Cement

The type of cement used in this work is (CMI / 42.5) from the plant of Amran-Yemen. It is 95% in clinker and 5 percent in gypsum. The chemical and mineralogical composition are determined by - X-ray Fluorescence (XRF) and their physical properties are presented in the tables (1, 2 and 3):

Table 1: Elementary chemical compositions of clinker, gypsum, and cement

Content (%)	CaO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	Mgo	SO ₃	K ₂ O	Na ₂ O	Cl
Clinker	62.76	21	5.84	3.00	1.96	0.9	1.21	0.2	0.02
Gypsum	33.4	0.7	0.36	0.09	0.63	47.2	0.03	0.1	0.01
Cement	61.29	19.99	5.57	2.85	1.89	3.22	1.15	0.2	0.02

Table 2: Mineralogical composition of clinker

Chemical name	Mineral name	Chemical formula	Cement nomenclature	Content
Tricalcium silicate	Alite	Ca ₃ SiO ₅	C ₃ S	47.7
Dicalcium silicate	Balite	Ca ₂ SiO ₄	C ₂ S	25.1
Aluminate tricalcium	Aluminate	Ca ₃ Al ₂ O ₆	C ₃ A	10.4
Tetracalcium Aluminoferrite	Ferrite	Ca ₄ AlFeO ₅	C ₄ AF	9.1

Table 3: Physical properties of clinker and cement

Physical properties	Units	Values	
		Clinker	3360
Specific surface Blaine	cm ² /g	Cement	3240
Absolute density	g/cm ³	Clinker	3.17
		Cement	3.14

2.1.2. Inorganic additions

2.1.2.1 Pure limestone (P, Lime)

It is a mineral material spread in several regions in Yemen, such as Hadramaut, Sana'a, and Amran, etc. This material with a volume about of 3.6 billion m³ [26, 27] (figure 1). The results of specific chemical analysis by the X-ray Fluorescence (XRF), mineralogical analysis by X-rays Diffraction (XDR) and the physics properties of the P. Lime of Bani Qais-Amran - Yemen after crushing, drying in the oven for 12 hours at 80 °C and grinding are shown in tables (4 and 5) and figure (3).



Figure 1: Deposit of extract the pure limestone in Bani Qais-Amran-Yemen

2.1.2.2 Natural pozzolan

The PN used is of volcanic origin, extracted from the deposit of Difan – Amran-Yemen (figure 2), located in the north of Sana'a [28, 29]. It consists essentially of slag and well-stratified pumice stones, color vary from red to black. The chemical analysis determined by X-rays Fluorescence (XRF), mineralogical analysis by X-rays Diffraction (XRD) and the physics properties after crushing and then steamed for 24 hours at a temperature of 50 °C in order to eliminate their moisture and grind until the resulting powder can pass through a sieve of 90 μm are shown in tables (4 and 5) and figure (4).



Figure 2: Deposit of extract the natural pozzolan in Difan-Amran-Yemen

Table 4: Elementary chemical compositions of P,Lime and PN determined by XRF

Content	CaO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	Mgo	SO ₃	K ₂ O	Na ₂ O	Cl	LOI
P,Lime	54.96	0.62	0.12	0.16	0.41	0.08	0.01	0.00	0.00	43.63
PN	8.8	41.43	16.16	9.41	4.79	0.13	0.90	3.47	0.04	14.87

From the results shown in the table (4) we found that the P,Lime of Bani Qais-Amran - Yemen contains 54.96% of lime (CaO) / 0.12 alumina (Al₂O₃) / 0.159 of iron (Fe₂O₃) / 0.621% of silica (SiO₂). In fact, the sum of the percentages is equal to 56.36% and the rest represents the loss on ignition (LOI) [30]. However, the PN from Difan-Amran contains 41.43% of silica (SiO₂) / 16.16% of alumina (Al₂O₃) / 9.41% of iron (Fe₂O₃) / 8.8% of lime (CaO). These percentages represent an amount equal to 85.13% and the rest as the loss on ignition [31].

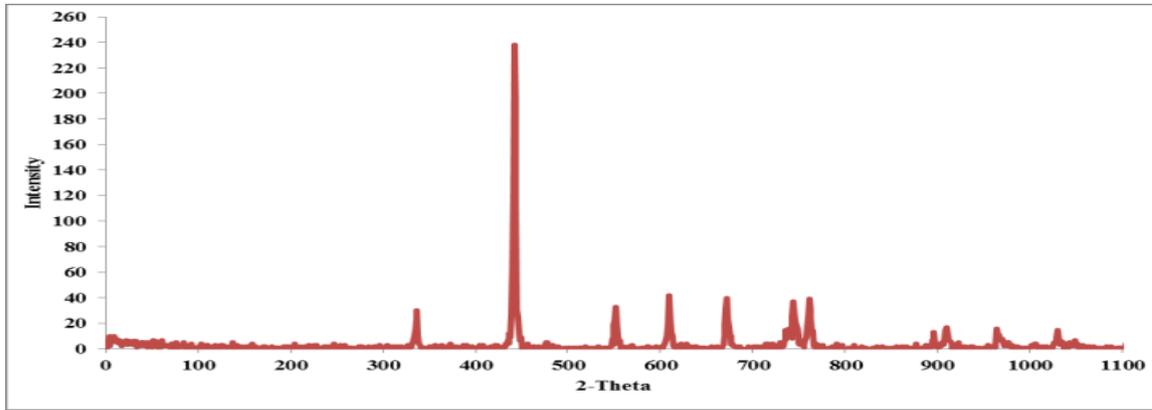


Figure 3: Spectrum of P,Lime

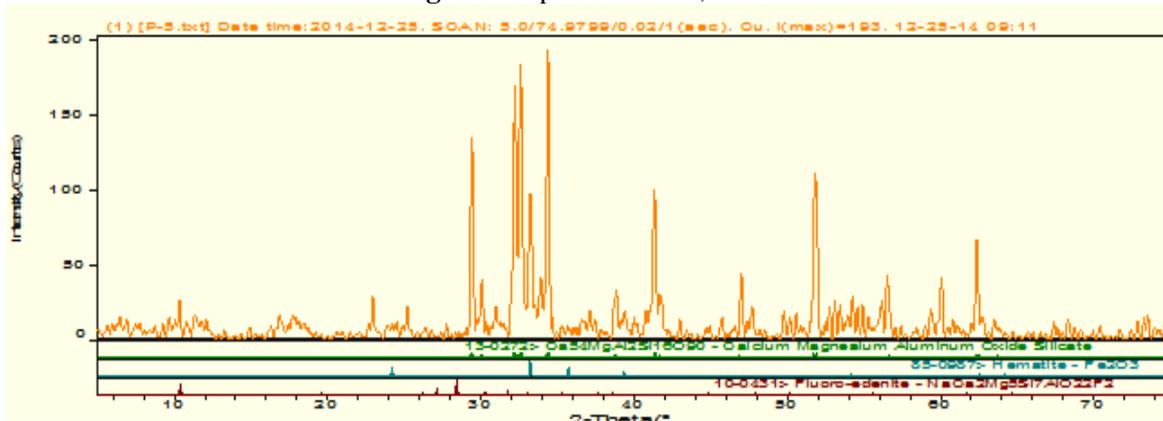


Figure 4: Spectrum of PN

According to the figures (3 and 4) that illustrate the mineralogical analysis determined by XRD of P,Lime and PN, we deduced the following comments:

- The P,Lime present in figure (3) reveals the strong presence of calcite (CaCO₃) then by the dolomite (CaCO₃/MgCO₃) as well as the magnesium carbonate (MgCO₃) and magnesium hydroxide Ca(OH)₂(OH)₂/Mg [32].
- The NP shown in figure (4) exposes the strong presence of the Feldspar, plagioclase (Anorthite: CaOAl₂O₃ 2SiO₂) followed by pyroxene (Augite: (Mg, Fe)₂ 2SiO₆) then the volcanic glass to Analcime (zeolite) Chlorite: (6 Mg₅ AlSi₃ O₁₀) (OH) as a result of Hematite: Fe₂ O₃ also as the Magnetite: Fe₂ O₃

Fe O as well as the Biotite: $2 K (Fe, Mg)_3 AlSiO_{10} (OH)$ and traces of minerals: basalts, dolomites, calcite, clays, etc. [33, 34].

The metallographic microscope of P,Lime and PN analysis gives an indication on the rearrangement of the particles of a composite state solid powder as shown in the figure (5, 6).

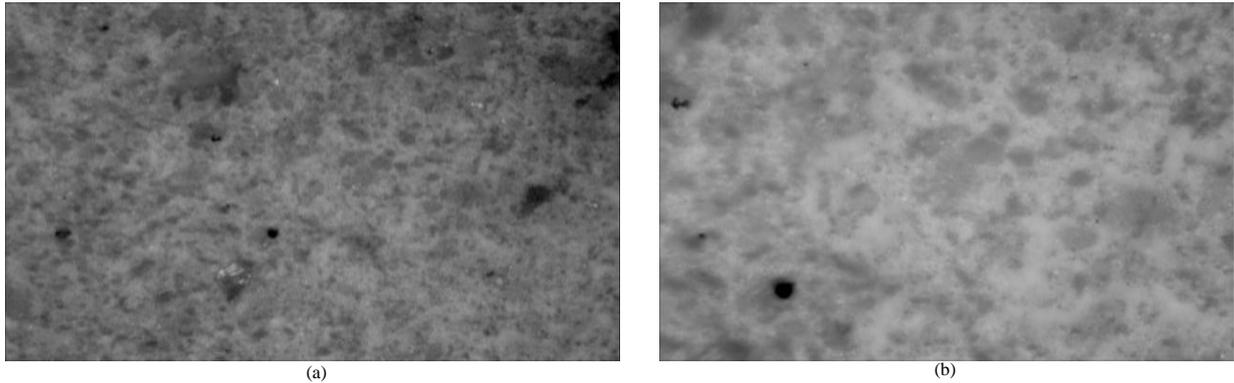


Figure 5: View by metallographies microscopic of P,Lime at 100 x (a) and (b) 200 x

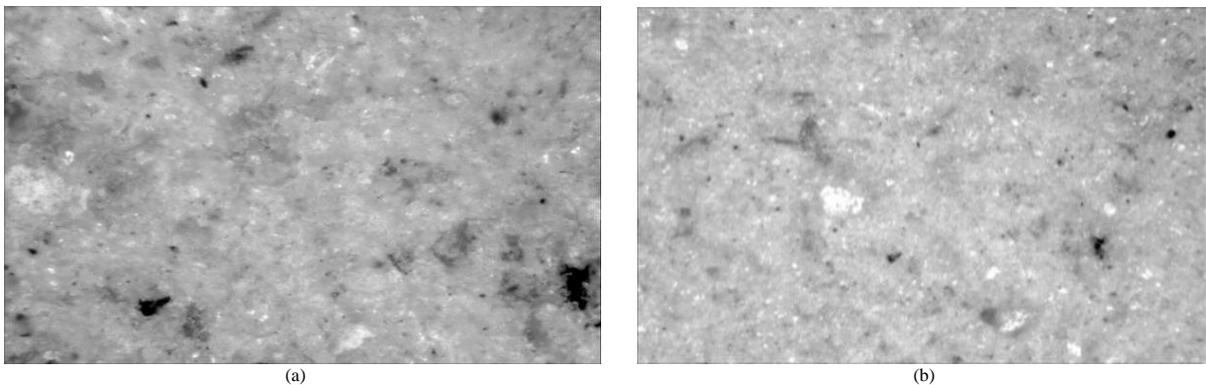


Figure 6: View by metallographies microscopic of PN at 100 x and 200 x (b)

The particles of the P,Lime and the PN are presented in the form of rosettes. They appear in the figures (5 and 6) using metallographies microscope to two expansions (100 and 200).

The physical properties

The physical properties of P,Lime and PN are given in table (5):

Table 5: Physical properties of P,Lime and the PN

Physical characteristics	Units	Values	
		P,Lime	PN
Specific surface Blaine	cm^2/g	4776	4576
Density	g/cm^3	2.13	2.81

2.1.3. The mixing water

To waste our mixture, we used tap water (wells), its main features are gathered at the table (6).

Table 6: Main features of the mixing water

Components	PH	T, D, N	CO_3^{-2}	HCO_3^-	Calcium	Magnesium (Mg^{+2})	Conductivity
Units	-	(mg/l)	(mg/l)	(mg/l)	(mg/l)	(mg/l)	$\mu S/cm$
Values	7.0	450	216	0.0	56.4	52.4	692.0

2.1.4. Sand

To prepare our mortar, we have used a standard sand conferring to the norm EN 196-1, delivered by the new French company of Littoral. Its particle size analysis is illustrated in figure (7).

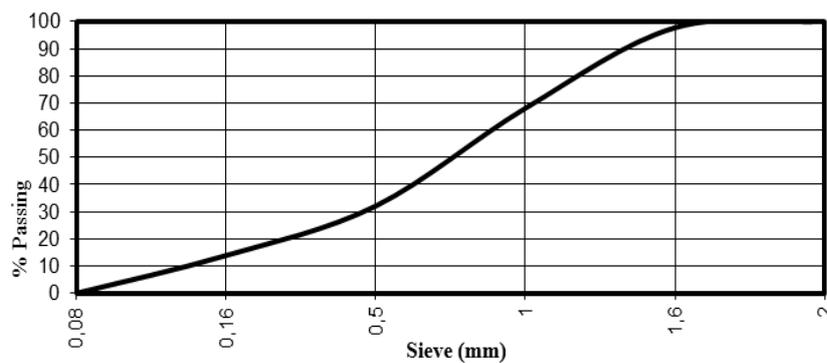


Figure 7: Grading Curve of sand

The particle size analysis presented in figure (7) there is that used sand grains are distributed in a systematic way according to the standard EN 196-1 specifications [35].

2.1.5. Software used

To determine the parameters affecting the compressive strength at 28 days, this represents the model of rupture, all the factors influencing and their interactions the module of the breaker was studied. A matrix of the Doehlert was established and processing the results with the Nemrodw software (New Efficient Methodology for Research using Optimal Design) enabled us to quantify the direct effects and interactions of the various factors on the responses chosen and to identify those that will require fine-tuning during the optimization phase.

2.2. Methods

2.2.1 Experimental protocol

We studied the influence of the combination of two inorganic additions, the PN and the P,Lime at different percentages ranging from 5% to 40% by weight of clinker with a step of 5% on the mechanical properties. Different formulations have been prepared to determine the compressive strength of the prismatic specimens of size $(4 \times 4 \times 16) \text{ cm}^3$. These specimens have been taken from a plastic mortar with a paste of standard consistency, according to the specifications of the norm EN 196 - 1 [36, 37].

2.2.2 Methodology of experimental design

The methodology that is conventionally used to study the influence of operating parameters on a The studied response is to change the value of a parameter while other fixed now [38]. The exploitation of results and the experimental study can be greatly simplified by using the methodology of the experience design [39, 40]. This technique allowed us to create a statistically significant model of a phenomenon that incorporates the interactions between the variables while optimizing the number of tests completed [41, 42]. When a hydraulic binder is influenced by three independent variables, a three-dimensional space was created in which the operating points can be defined. All values obtained during the measurement of the compressive strength are signals of the response of these three-dimensional operating points.

Response surface methodology (RSM)

The RSM which is an empirical statistical technique used of multiple regression analysis with quantitative data for statistical experiments designed, will solve the equations of multivariable simultaneously. The graphic representation of these equations is called response surfaces [43, 44]. It allows to describe the impact of the individual and cumulative test on the response variables and determine the mutual interaction between them. The main objective of the RSM is to determine the optimal and operating conditions for a given system that satisfies the operating conditions.

Multiple linear regression models (MLRM)

The MLRM is the most current statistical tool for the study of multidimensional [45, 46].

2.2.3. Determination of the factors

The optimization of the formulation matrix of a new hydraulic binder at the base of a combination of PN and P,Lime is based on three factors that influence the compressive strength at 28 days. We chose a matrix of the

Doehlert experience that allows finding optimal qualities predicting the response calculated at all points in the field. Its various factors, coded variables and the matrix of experimental design are collected in the tables (7, 8 and 9).

Table 7: The experimental field of the factors studied

Factors	Notation	Quantity		Unity
		Low level (-1)	High level (+1)	
PN	X1	0	40	%
P,Lime	X2	0	40	%
Report W/C	X3	0.4	0.54	%

Table 8: Coded Variables

N°Exp	X1	X2	X3
1	-1	-1	-1
2	1	-1	-1
3	-1	1	-1
4	1	1	-1
5	-1	-1	1
6	1	-1	1
7	-1	1	1
8	1	1	1
9	-1	0	0
10	1	0	0
11	0	-1	0
12	0	1	0
13	0	0	-1
14	0	0	1
15	0	0	0

Table 9: Matrix of experimental design

N°Exp	Rand	Pozzolan	Pure Limestone	Report
		%	%	W/C
1	9	0	0	0.4
2	1	40	0	0.4
3	14	0	40	0.4
4	10	40	40	0.4
5	4	0	0	0.54
6	12	40	0	0.54
7	11	0	40	0.54
8	5	40	40	0.54
9	8	0	20	0.47
10	3	40	20	0.47
11	15	20	0	0.47
12	6	20	40	0.47
13	13	20	20	0.4
14	7	20	20	0.54
15	2	20	20	0.47

3. Statistical analysis

3.1. Equation of model:

For a detailed study of the formulation of our new hydraulic binder, we used a model of a second-degree polynomial (Equation.1). It includes linear effects, interaction effects, and quadratic effects of factors. The response surface model of the second degree can be written by the equation (1):

$$Y_i = \beta_0 + (\sum_{i=1}^n \beta_1 X_i^1 + \beta_2 X_i^2 + \dots + \beta_F X_i^P)^2 + \epsilon, \quad \text{Eq (1)}$$

From the equation (1), we replaced the variable x-factors and we will eventually get (2).

From equation (1), we have replaced the variables x by the factors and we will obtain a second-degree polynomial equation (2):

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_{11} X_1^2 + \beta_{22} X_2^2 + \beta_{33} X_3^2 + \beta_{12} (X_1 X_2) + \beta_{13} (X_1 X_3) + \beta_{23} (X_2 X_3) + \varepsilon, \text{ Eq (2)}$$

With:

Y: The response

X₁: The level attributed of the factor 1 (pozzolan proportion);

X₂: The level attributed of the factor 2 (P,Lime proportion);

X₃: The level attributed of the factor 3 (report W/C);

β₀: The value of the response of the center of the field study of response surface;

β₁: The effect of factor 1.

β₂: The effect factor 2.

β₃: The effect factor 3.

β₁₂: The interaction between factors 1 and 2;

β₁₃: The interaction between factors 1 and 3;

β₂₃: The interaction between the factors 2 and 3;

ε: The residues

3.2. Estimation and statistical coefficients

All the factors coefficients studied as well as their effects, then its statistical values (t-student) and the observed probability (Signif) are grouped in the table (10). The values of t-student is used to determine the significance of the coefficients of each factor, whereas the Signals are defined as the smallest significance level. In general, the larger of t-student size, the smallest of Significance, and the more significant the coefficient term [47, 48].

These statistical coefficients are:

- The constant: β₀;
- The linear terms: β₁/ β₂ and β₃;
- The quadratic terms: β₁₁/ β₂₂ and β₃₃;
- The interaction terms: β₁₂/ β₁₃ and β₂₃;

Table 10: Statistical estimation of the coefficients of the model associated with the compressive strength and their importance in relation with respect to the experimental dispersion

Coefficient	Estimated value	F.Inflation	Standard deviation	Value of t _{exp}	Significance
β ₀	38.250		0.574	66.66	< 0.01 -
β ₁	-0.917	1.00	0.338	-2.72	4.17 **
β ₂	0.794	1.00	0.338	2.35	6.5 NS
β ₃	-0.441	1.00	0.338	-1.31	24.8 NS
β ₁₁	-0.015	1.30	0.666	-0.02	98.1 NS
β ₂₂	0.390	1.30	0.666	0.59	58.7 NS
β ₃₃	-4.145	1.30	0.666	-6.23	0.204 **
β ₁₂	0.883	1.00	0.377	2.34	6.6 NS
β ₁₃	0.265	1.00	0.377	0.70	51.8 NS
β ₂₃	0.248	1.00	0.377	0.66	54.5 NS
** : Very significant coefficient			NS: not significant		

According to the table (10), which shows the different coefficients of the selected model that can be analyzed statistically, we found that there are low values of significance (often less than 5%) indicating that the model has a good coefficient significance, that is to say, had a confidence level of 99.9%.

The third column of the same table gives the inflation factor which is an absolute measure of the independence of the coefficients [49, 50], in other words, it measures the degree of orthogonality of the matrix of experiments. This shows that the quality of information decreases with increasing this factor. In other ways, an experiment matrix provides desired information if the inflation factor the closer to 1.

3.3. Analysis of results

3.3.1. Analysis of variance

The table (11) displays the results of the analysis of variance, significant and the level of confidence of the model obtained.

The presented results in the table (11) indicate that the main effect of the regression is significant since the probability of the significance of the p-value risk is less than 5%, hence the model is statistically at a significant good, especially as it has a higher confidence level of 95% [51, 52].

Table 11: Analysis of variance of the obtained model

Source of variation	Sum of squares	Degrees of freedom	Average square	Report	Significance
Regression	77.5576	9	8.6175	7.5606	2.10 *
Residues	5.6989	5	1.1398	-	-
Total	83.2565	14	-	-	-

3.3.2. Statistical analysis of results

In order to judge the quality of the model chosen using a numerical indicator, we limit to use the multiple linear correlation coefficient R^2 . It is well known that this coefficient must be handled with caution, so it is more prudent to accompany it in practice by calculating another coefficient such as the adjusted multiple linear correlation coefficient R^{2a} [53, 54].

The tables (12 and 13) present the comparisons between the experimental results and the values calculated from the postulated model and their quality of validation.

Table 12: Comparison between experimental results and the values calculated from the postulated model

N°Exp	Yexp	Ycal	Residue
1	36.450	36.439	0.011
2	32.150	32.310	-0.160
3	35.150	35.767	-0.617
4	35.320	35.168	0.152
5	34.530	34.532	-0.002
6	32.230	31.463	0.767
7	35.160	34.850	0.310
8	35.450	35.311	0.139
9	39.450	39.152	0.298
10	36.420	37.318	-0.898
11	37.230	37.846	-0.616
12	39.450	39.434	0.016
13	35.160	34.546	0.614
14	32.450	33.664	-1.214
15	39.450	38.250	1.200
	Exp: experimental	cal: calculated	Residue: $y_{exp} - y_{cal}$

Table 13. Estimates and statistics of the coefficients of the model applied

Standard deviation of response	1.068
R^2	0.932
R^{2a}	0.808
R^2 pred	0.445
PRESS	46.231
Number of degrees of freedom	5
PRESS: Predicted Residual Sum of Squares	

From the table (13), which shows the estimation and statistics of the coefficients of postulated model, we observed that all responses have satisfactory descriptive qualities because the correlation coefficient $R^2 = 0.932$ and the adjustment coefficient $R^{2a} = 0.808$ are sufficient since they have values closer to 1. These values give better compatibility between the experimental and predicted values of the adapted model. The figure (7) shows that the predicted values are close to the experimental ones.

3.3.3. Statistical analysis of residues:

To confirm that the model describes well the variation in responses, it was necessary to ensure that locally, the residues are not high (Figure 8). The normality of the distribution of the residues is an important assumption of the method of least squares method. Considering the number of tests present in an experimental design, we generally use the Henry graphic method (Figure 9).

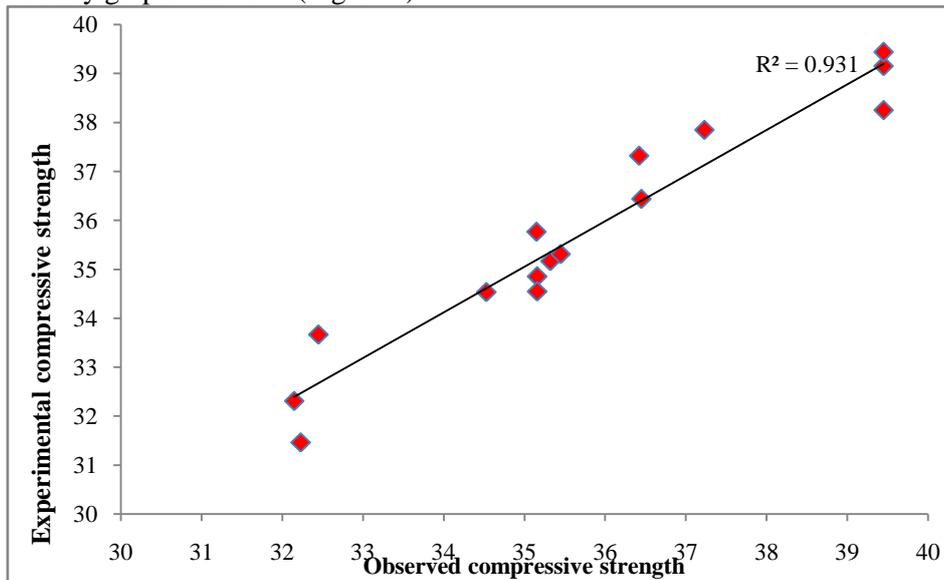


Figure 8: Observed values as a function of the expected values of the rupture model of compressive strength

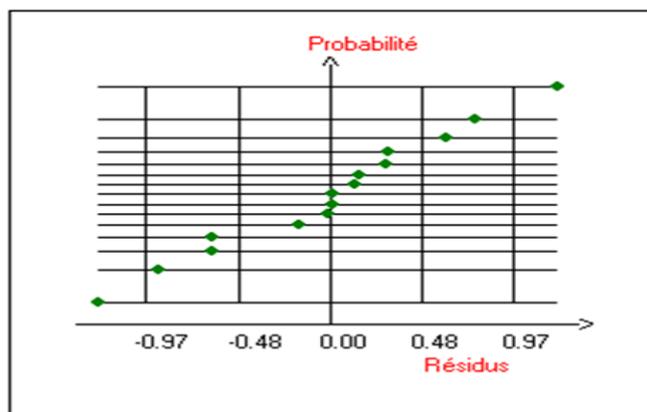


Figure 9: Right of Henry

From the figure (9), we have observed that the cloud is close to a straight line, that is to say, that, the distribution of the real values of the variable is normal, because the linearity is satisfactory.

4. Results and discussion

4.1. Optimization:

The aim of the optimization is to answer a specific objective in our study, which is the formulation of a new durable hydraulic binder based on different percentages of inorganic additions: PN and P,Lime, all partially substituted the cement by these additions at various percentages ranging from 5% to 40% by weight of the cement with a step of 5%. It's about finding the operational values of the factors, which provoke the desired response by relying on economic and environmental constraints. After the system analysis and the modeled response based on the three factors, the optimum of this new formulation can be shown by the iso-response method curves

4.1.1. Response surface and iso-response

The response surface represents the regression surface from a graph in a three-dimensional space [54, 55]. It schematics on the horizontal plane the domain of the variation of 2 factors; The vertical axis shows the variation of the response from the model. Beyond 2 factors, it is necessary to maintain at a constant level. However, the

The mentioned results in the table (15) show that there is no significant difference between the experimental and predicted responses.

5. Conclusion

The objective of this work is the formulation of a new durable hydraulic Binder prepared from an inorganic additions: the natural pozzolan (PN) and the pure limestone (P,Lime) in partial substitutions the clinker by these additions at different percentages ranging from 5% to 40% by weight of cement with a step of 5%. To achieve this objective, we have optimized the factors (the PN content, the P,Lime content and the report W/C). In order to improve the physical-chemical and to obtain a mathematical model that predicts the compressive strength, while using a methodology of experimental design, more precisely the approach of the response surface, we also modeled and optimized the compressive strength at 28 days which represents in our case the model of the rupture of our new hydraulic binder.

According to the mathematical model obtained, the compressive strength depends on the linear terms β_1 , relative to the proportions of PN and the quadratic terms β_{33} , relative to the report W/C.

After the validation of the model obtained, we observed that the experimental and optimal conditions to maximize the compressive strength are the following: the PN content is of the order of 35 % with a report W/C is 0,42.

These results have enabled us to develop a new durable hydraulic binder with improved physical-chemical and mechanical properties on one hand and on the other hand, is represented a great economic and environmental interest:

- Reduction of the production costs;
- Minimizing the emissions of CO₂ into the atmosphere;
- Decrease the use of mixing water.

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