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Optimization of a Self-Compacting Paste Based on Glass Powder Using Mixture Design and the Desirability Function

Messaouda Ch. Boulkhiout[1*], Ryad Amdoun[2], El Hadi Benyoussef[1], Abderrahim Bali[1]

[1] Laboratory of Civil Engineering Materials and Environment, Ecole Nationale Polytechnique, Algiers, Algeria
[2] Institut National de la Recherche Forestière (INRF), Baïnem - BP 37, Cheraga, Algiers, Algeria
*e-mail: messaouda.cherrak@g.enp.edu.dz

Abstract

The partial replacement of cement by supplementary cementitious materials is an opportunity for reducing fossil energy consumption and greenhouse gas emissions. In this study, forty (40) pastes were formulated by applying a 4-factor mixture plan, consisting of cement, glass powder (GP), superplasticizer and water. Third order models "special cubic" established for the flow time, spreading diameter, and flow velocity, responses were statistically significant. The analysis of the models allowed the study of the individual and combined effects of the different factors, highlighting the predominant effect of the superplasticizer on the dispersion GP fines. The multi-criteria optimization allowed the development of two optimal pastes SCP1 and SCP2 incorporating 25 and 7% GP, respectively. SCP1 was chosen because at this GP amount, unlike at 28 days, the paste presents the best strengths at 90 days with 4.5% increase for flexural tensile strength and 15.69% increase for compressive strength while maintaining satisfactory workability.

Keywords: desirability function, self-compacting paste, mixture design, glass powder, fresh properties, recycling, environment, compressive strength

1 Introduction

Glass waste produced in urban areas causes major environmental problems with low recycling rates. The annual global production of glass is about 130 million tons. Despite efforts to recover glass, only 27 million tons have been recovered worldwide, representing 1/5 of the total glass waste produced [1]. These figures show that efforts still need to be made to recycle this non-biodegradable waste. It takes a million years to decompose. In order to reduce urban environmental problems, glass waste is recycled and valorized in the construction industry, notably cement and concrete [2].

Self-compacting concrete (SCC) is an innovative workable material that is characterized by its high capacity to set up under its own weight without vibration or compaction. SCC made of only with cement can lead to high costs and other technical problems such as higher hydration heat. Therefore, Supplementary Cementitious Materials (SCMs) are frequently used as a partial cement replacement, reducing heat of hydration, increasing compactness, and reducing

segregation while maintaining appreciable workability [3, 4]. In addition, using these cementitious materials helps save on cement quantities and reduces the carbon footprint.
In this context, since the 2000s, several researchers have worked on the valorization of glass in concrete industry and SCC in general, and in pastes in particular, notably by using it either as a supplementary cementitious material or as an addition [5 - 7]. More extensive research is needed to overcome the barrier of the specific percentage of GP contribution in fresh and hardened properties [8].
The research focused on the study of the effect of Glass Powder (GP) on the various performances of concrete, mortar or paste. Up to 30% cement, replacement by GP shows no major effect on mechanical performance. In fact, 30% GP could be incorporated into the concrete without any harmful long-term effect. Examination of the mortar samples at 270 days showed a dense microstructure thanks to the pozzolanic reaction of the fine glass particles. Despite the lower initial strengths, the concrete samples continued to develop strength over time and approached the strength of the control mix [9, 10]. Regarding rheological behavior, different opinions are put forward. Hence, the increase in workability as a function of the percentage of GP has been investigated, in particular, for SCC. An increase was observed up to 20% substitution, then the trend reversed and a drop in fresh state performance was recorded. This trend was attributed to the low water absorption and smooth surface of GP [11, 12]. In addition, other research has shown the opposite effect with a loss of fluidity. This loss, which was accentuated by the in the percentage of GP substitution, was attributed to the low density of GP which is less important than that of cement. An equal mass substitution of cement by GP increases the friction between solid particles in the paste by increasing the solid/water volume ratio [13, 14].
The complexity of incorporating crushed glass in concrete technology requires further research to confirm the effectiveness and conditions of use of this material as an alternative cementitious material [15]. Clear answers are needed in the near future to clear up the existing ambiguities caused by unclear trends in the effect of GP on the fluidity especially of cementitious mixes. With regard to future research trends in this area of glass waste recovery, they are moving towards high substitution dosages with possible recourse to activators in order to achieve the expected performance with low quantities of cement. Efforts are still being made to protect the environment and manufacture low-carbon materials.
The present study is a part of this area of research where the aim is to investigate the effect of recycled GP from bottles on the workability of self-compacting pastes (SCP). The main objective is to study the influence of PV on cement paste fluidity, focusing on the individual effects and possible interaction of the various inputs. To carry out this study, the mixing experiments plans, thanks to a reduced number of experiments, allow the construction of mathematical models explaining the variation of the responses according to the materials proportions in the mix. The adopted plan was built by considering the various components of the paste as factors: cement, glass powder, superplasticizer, and water. The third-order "special cubic" models established for the responses flow time, T(s); spreading diameter, D(cm) and fluidity velocity D/T (cm/s), are established and analyzed.
The determination of the optimal mix proportions with complex responses requires the simultaneous consideration of several responses. It is therefore a case of a problem based on multi-criteria decision making [16-18]. Satisfactory solutions can be obtained by applying the desirability function [19, 16].

The combined application of a mixture design and the desirability function results in a more powerful approach to design an optimal GP-based self-placing paste.

2 Materials and Methods

2.1 Materials Used

Self-compacting pastes are prepared using 42.5 N Portland cement (Type II), a superplasticizer (SP) and a glass powder (GP) whose physical characteristics are summarized in Table 1. The GP is obtained from recycled bottles, crushed for 15 hours, and then sieved.

Table 1: Physical characteristics of the materials used

Portland cement CEM II	**Superplasticizer**	**Glass powder**
Standard : NF EN 197-1 [16]	Type : polycarboxylate	Bulk density : 2500 kg/m^3
Bulk density : 3060 kg/m^3	Aspect : Liquid	Fineness (BSS) : 2800 cm^2/g
Fineness (BSS) : 3700 cm^2/g	Color : Brown	Size grading $\leq$ 63µm
Setting time : 60 mn	PH : 9.5-10.5	
Compressive strength: 50 MPa	Density : 1.19 ± 0.01	

Glass powder (GP) which is rich in silica as shown in (Table 2), constitutes a main element in the improvement of the pozzolanic reaction.

Table 2: Chemical characteristics of cement and glass powder (GP)

Constituents	**SiO_2**	**Al_2O_3**	**Fe_2O_3**	**CaO**	**MgO**	**SO_3**	**K_2O**	**Na_2O**	**CrO_3**
Cement	21.31	3.00	3.29	65.39	3.74	0.66	0.72	0.18	-
GP	71.12	1.71	0.24	10.02	3.01	0.25	0.19	13.1	0.23

2.2 Mixing Procedure

The preparation of self-compacting pastes consists of mixing first the liquids, water, and SP at low speed for 1 min. Then, the paste is elaborated by adding solid suspensions from cement and GP which were beforehand dry homogenized. The solid fraction is introduced gradually for 2 mn mixing at low speed then for 1 mn at high-speed meeting the recommendations of Maher [21].

2.3 Mixture Design

Preliminary tests were carried out to delimit the study domain for obtaining workable pastes, neither too firm nor too liquid and not showing obvious signs of segregation [21]. The defined study domain is a 4-dimensional space, and the levels of variation of the studied factors (constituents) and the position of the candidate points for the determination of the experiment matrix are reported in Table 3.

Table 3: Levels of the studied factors, position, and number of points in the experiment matrix

Mixture design factors			**Mixture design points**	
	Min	**Max**	**Position**	**Number**
C : Cement	0.43	0.550	Center	5
GP : Glass powder	0.00	0.130	Edge	8
SP : Superplasticizer	0.00	0.026	Interior	8
W : Water	0.38	0.500	Plane	14
			Vertex	5
			Total (matrix)	40

The generated mixture plan consists of 40 points (mixtures or experiments). The results from the application of this plan will be used to fit the third order models for each measured response.

2.4 Measured Responses

A self-compacting paste must be stable, homogeneous, and fluid. To characterize the rheological activity of the self-compacting paste, four responses are measured: flow time *T*(s), paste spreading diameter *D*(cm), paste segregation Seg and flow velocity *D/T* (cm/s).

2.4.1 Flow Time *T* (s)

The flow time in seconds, *T* (s), is evaluated by the standardized Marsh cone test according to the (NF P18 507) standard [22]. Its purpose is to characterize the flow of self-compacting paste and its ability to fill a fixed volume by measuring the time required in seconds, *T* (s), to fill a one-liter container. It is a simple and practical method for obtaining a relative measure of the flowability of a paste.

2.4.2 The Paste Spreading *D* (cm)

The paste spreading diameter *D* (cm), is measured by the mini-slump cone which is first filled and then lifted allowing the cement paste mass to spread. Once stable, the average diameter of the cake is recorded. The mini-cone test or spreading test is inspired from the Abrams cone test but with its appropriate dimensions as shown in Figure 1 [23]. This test consists of filling the mini-cone placed on a horizontal surface and measuring the diameter of the obtained spread paste in both directions. After raising the cone and waiting for 1 minute, the diameter value will be the average of the two recorded measurements. Thus, it gives an indication on the workability of the elaborated flowing paste.

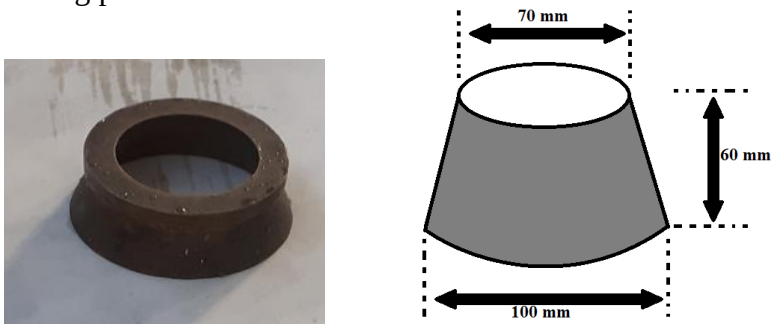


Figure 1: Mini cone for spreading test

2.4.3 Segregation of the Pastes

The segregation of the pastes has been also assessed by considering the visual aspect and the bleeding water on the surface after a settling time of 2 hours. Pastes with a risen water level not exceeding 2 mm are considered stable without segregation and designated by the value 0. However, when an excessive bleeding water level occurs and exceeds a height of 2 mm, the pastes are considered unstable and result in segregation. In this case the value of 1 is assigned to the Seg response.

2.4.4 The Flow Velocity *D/T* (cm/s)

This last parameter is the ratio between the first two responses (*D/T*) in order to characterize the rheology in terms of the flow velocity of the self-locking paste (cm/s).

2.5 Statistical Modeling

In the case of mixture, the constituents (factors) are dependent on each other. Classical experimental design methods such as factorial design are based on the independence of the factors. For this reason, they are not applicable for the development of cement paste formulation model performed at constant volume where the sum of the proportions of all constituents is equal to 1 or 100%. Thus, the mixture design is chosen. The measured responses depend on the proportions of the components [24, 25]. The mixture is composed of q components where x_i represents the proportion of the i^{th} component. The following equation can be written as follows:

$$0 \leq x_i \leq 1 \quad , \mathrm{i} = 1, 2, \ldots . q \tag{1}$$

$$\sum_{i=1}^{q} x_i = 1 \tag{2}$$

In the case of stresses on the variables, Equation (1) becomes:

$$0 \;\leq\; \mathrm{L_i} \leq x_i \leq \mathrm{Ui} \;\leq 1 \quad , \quad i = 1, 2, \ldots . \mathrm{q} \tag{3}$$

where, *Li* and *Ui* are the lower and upper stresses of the i^{th} component respectively. The applied stresses reduce the experimental domain to an irregular dimensional space *(q - 1)* called irregular hyper-polyhedron.
The third order polynomial model "special cubic" is chosen for this study (Equation 4).

$$Y = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j} \sum \beta_{ij} x_i x_j \; + \sum_{i<j} \sum_{j<k} \sum \beta_{ijk} x_i x_i \, x_k \tag{4}$$

where β_i, β_{ij} ,β_{ijk} are the coefficients of the regression equation calculated by the least squares method (Goupy and Creighton 2006). x_i, x_j and x_k are the proportions of the i^{th}, j^{th} and k^{th} component of the mixture respectively.

2.6 Statistical Analysis and Diagnosis

In order to assess the relevance of the model to perfectly describe the studied domain, it is necessary to calculate the fit statistics, to make a diagnosis and an ANOVA of the model.
The fit statistics that will be taken into account are the coefficient of determination R^2, the R^2adj and the R^2pred. Values of R^2 close to 1 indicate the adequacy of the model with the variability of the data collected. R^2 increases with the number of variables even if they are not statistically significant [18], so it is necessary to consider the R^2adj which decreases when the number of non-significant terms in the model increases. The R^2pred measures the amount of variation in the new data predicted by the model and should not differ from R^2 by more than 0.2. A large deviation would indicate a problem in the data or in the model. In this case, several possibilities can be taken into account in the diagnosis of the regression: - verification by graphical methods of the Normal Plot of Residuals, - the possibility of the Box-Cox Plot for Power Transforms, - the search for outliers, - the choice of another model or its modification by deleting statistically non-significant terms.
The statistical significance of the model, the factors and their interactions is determined by analysis of variance (ANOVA, Fisher F-value test).
If the fit statistics, ANOVA and diagnosis are satisfactory, the resulting model can be used in the study and the optimization. These statistical analyses were carried out using the Design Expert 13 software demonstration version to draw the different diagrams and proceed with the optimization.

2.7 Multiresponse Optimization by Desirability Function

Desirability is based on the transformation of all obtained responses *Yi* into a desirability function *di(Yi)* from different scales into a scale-free value ranging from 0 to 1 [16, 19]. The value 0 is assigned when the factors give an undesirable response and the value 1 to the optimal performance for the factors under consideration. Two types of transformations are considered according to the objectives: the unilateral transformation in the case of maximization or minimization of *Yi*, and the bilateral transformation to reach a target value t_i for *Yi* (Equation 5).

$$d_i(Y_i) = \begin{cases} 0, & Y_i < l_i \\ \left(\frac{Y_i - l_i}{t_i - l_i}\right)^s, & l_i \le Y_i \le t_i \\ \left(\frac{Y_i - u_i}{t_i - u_i}\right)^t, & t_i \le Y_i \le u_i \\ 0, & Y_i > u_i \end{cases} \quad (5)$$

where, l_i and u_i are respectively the lower and upper bounds of the studied response ($l_i < t_i < u_i$). The *s* and *t* correspond to the weighting of the factor depending on the user according to the sought degree of desirability for a given response between l_i and u_i. The overall desirability

function (D) is the geometric mean of the individual desirability values of each response $d_i(Y_i)$ calculated by Equation 6:

$$D = (\prod_{i=1}^{n} d_i(Y_i))^{\frac{1}{n}} \quad (6)$$

In order to achieve the desired workability, the T(s) response is minimized, and the spread D(cm) and D/T (cm/s) responses are maximized to obtain pastes with good workability and zero segregation (Seg).

2.8 Validation of Model

The comparison between the predicted and measured values by the *t-test* has been conducted in order to judge the validity of the multi-response optimization. The t-test compares the experimental mean m with the calculated theoretical mean μ according to Equation 7 [26].

$$|t| = \frac{m-\mu}{s/\sqrt{n}} \quad \text{n} \quad (7)$$

where, m is the observed mean, μ the theoretical mean, s is the estimated standard deviation of the sample and n the number of repetitions. If $|t| < 4.303$ at 5% risk, the difference is not significant then the model is validated. If $|t| > 4.303$, the difference is significant, so the model is non-validated.

2.9 Mechanical Strength of Optimal Pastes

Compressive and flexural strengths have been assessed only for the optimal pastes, in order to decide on the final formula to be used which should meet all the workability and strength criteria.
The strength measurements were carried out on 40 × 40 × 160 mm prismatic specimens meeting the (EN 1015-11) standard [27]. The tests were conducted at 28 days and 90 days of curing. Three specimens were prepared for each mix to monitor the variability of the different cement paste design. Each test result reported represented the average strength of three test samples.

3 Results and Discussion

3.1 Mixture Design Results and Responses Modeling

The results of application of the mixture design are summarized in Table 4.
For each response, a special cubic model was fitted to the mixture design results (Table 4). The coefficients of the fitted models for the four responses are summarized in Table 5.
Prior to the exploitation of these models, a diagnosis of the regression should be made to check the normal distribution assumptions of the residues and to detect possible "outliers". After ANOVA, the modification of the models by deleting insignificant terms is also considered in order to improve the relevance of the models. The deletion of these terms should lead to an improvement in the values of R^2, *Adjusted* R^2, *Predicted* R^2 and *Adeq Precision*.

Table 4: Mixture design results

Run	**Factors**					**Responses**			
	C	**GP**	**SP**	**W**	**%GP**	***T*(s)**	***D*(cm)**	****Se***	***D*/*T***
1	0.467	0.036	0.02	0.476	6	9.45	42.38	1	4.48
2	0.430	0.103	0.00	0.460	16	39.5	32.88	0	0.83
3	0.550	0.044	0.02	0.380	6	242.	35.13	0	0.15
4	0.487	0.062	0.01	0.437	9	27.8	39.00	0	1.40
5	0.462	0.101	0.02	0.415	15	31.6	42.38	0	1.34
6	0.453	0.073	0.00	0.467	12	20.6	31.13	0	1.51
7	0.461	0.099	0.00	0.440	15	100	10.00	0	0.01
8	0.549	0.071	0.00	0.380	10	100	10.00	0	0.01
9	0.430	0.083	0.02	0.461	14	10.3	41.00	1	3.97
10	0.467	0.036	0.02	0.476	6	8.46	44.25	1	5.23
11	0.430	0.130	0.01	0.422	20	30.0	41.50	0	1.38
12	0.453	0.073	0.00	0.467	12	19.6	31.75	0	1.62
13	0.487	0.062	0.01	0.437	9	22.6	39.50	0	1.75
14	0.492	0.120	0.00	0.380	17	100	14.00	0	0.01
15	0.487	0.062	0.01	0.437	9	23.0	39.50	0	1.72
16	0.501	0.068	0.02	0.405	10	34.8	45.00	1	1.29
17	0.519	0.000	0.01	0.462	0	15.3	42.00	1	2.74
18	0.487	0.062	0.01	0.437	9	23.9	38.50	0	1.61
19	0.430	0.070	0.00	0.500	12	100	10.00	0	0.01
20	0.487	0.062	0.01	0.437	9	24.1	40.00	0	1.66
21	0.550	0.030	0.01	0.408	4	67.0	31.50	0	0.47
22	0.456	0.077	0.02	0.441	12	17.7	40.50	1	2.28
23	0.550	0.000	0.02	0.424	0	27.3	40.00	1	1.46
24	0.519	0.000	0.01	0.462	0	13.3	46.00	1	3.46
25	0.550	0.000	0.00	0.450	0	100	10.00	0	0.01
26	0.527	0.050	0.00	0.419	7	100	13.50	0	0.01
27	0.511	0.040	0.02	0.423	6	25.3	40.75	1	1.61
28	0.517	0.083	0.02	0.380	12	50.5	37.50	1	0.74
29	0.464	0.102	0.01	0.414	15	32.5	40.50	1	1.24
30	0.464	0.130	0.02	0.380	19	48.4	39.00	0	0.81
31	0.454	0.130	0.00	0.416	19	100	10.00	0	0.01
32	0.491	0.032	0.00	0.477	5	100	10.00	0	0.01
33	0.492	0.120	0.00	0.380	17	100	12.00	0	0.01
34	0.460	0.039	0.00	0.500	6	9.38	32.50	0	3.46
35	0.470	0.004	0.02	0.500	1	4.82	43.00	1	8.92
36	0.527	0.050	0.00	0.419	7	100	13.00	0	0.01
37	0.499	0.088	0.00	0.413	13	100	10.00	0	0.01
38	0.446	0.028	0.02	0.500	5	4.88	44.50	1	9.12
39	0.430	0.054	0.01	0.500	9	5.72	45.50	1	7.95
40	0.493	0.000	0.00	0.500	0	8.20	34.50	0	4.21

Table 5: Estimated coefficients of fitted models

Full model terms	**Responses**			
	***T*(s)**	***D*(cm)**	**Seg**	***D*/*T*(cm/s)**
Cement	499.93	20.41	-0.29	2.34
GP	1159.72	6.89	0.21	3.21
SP	59516.04	-1468.75	63.66	-96.26
Water	-524.24	45.44	0.31	11.15
Cement – GP	2455.52	-56.13	1.10	-12.86
Cement – SP	-73340.42	1879.87	-74.20	114.49
Cement - Water	3718.79	-85.83	0.18	-25.08
GP – SP	-78639.86	2016.45	-80.11	105.30
GP – Water	1561.91	-43.69	-0.82	-30.69
SP – Water	-64337.30	1723.24	-73.27	186.14
Cement - GP - SP	-2644.89	160.16	19.97	6.03
Cement - GP - Water	-8650.25	202.41	-8.94	52.86
Cement - SP - Water	-24877.91	727.66	53.75	-82.41
GP - SP - Water	-9172.96	325.12	54.26	-9.29

3.2 Regression Diagnostic and Model Modification

Two graphic diagnoses are considered: the "Normal Plot of Residuals" (Figure 2) and the "Residuals vs. Runs" (Figure 3) to detect possible “outliers” values.

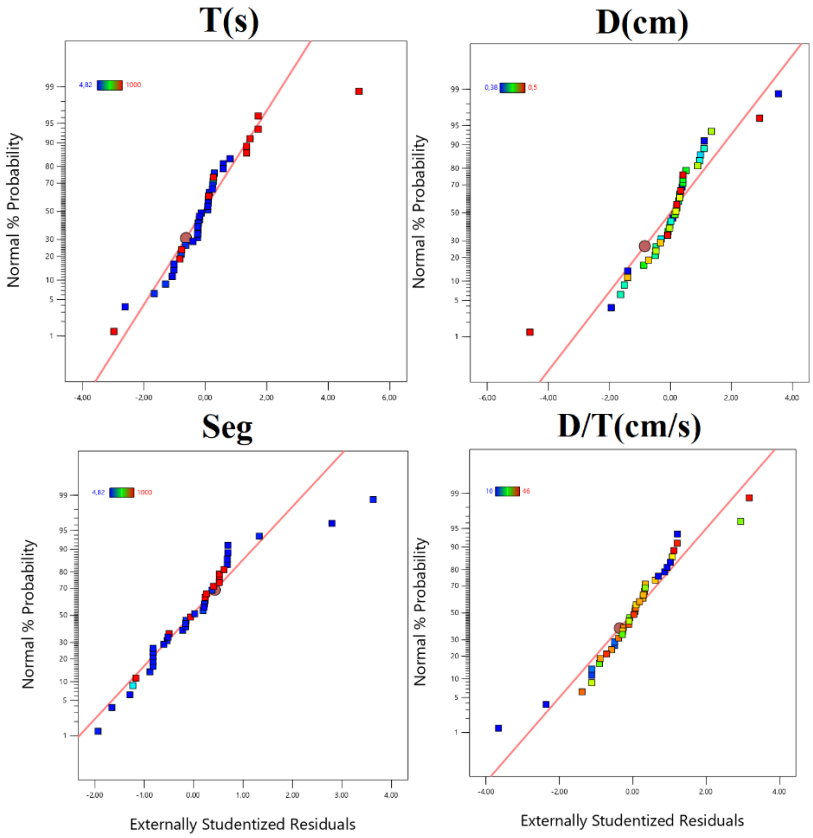


Figure 2: Normal Plot of Residuals. *T*(s): flow time. *D*(cm): spread of paste. Seg: segregation of the pastes. *D*/*T*(cm/s): Flow speed.

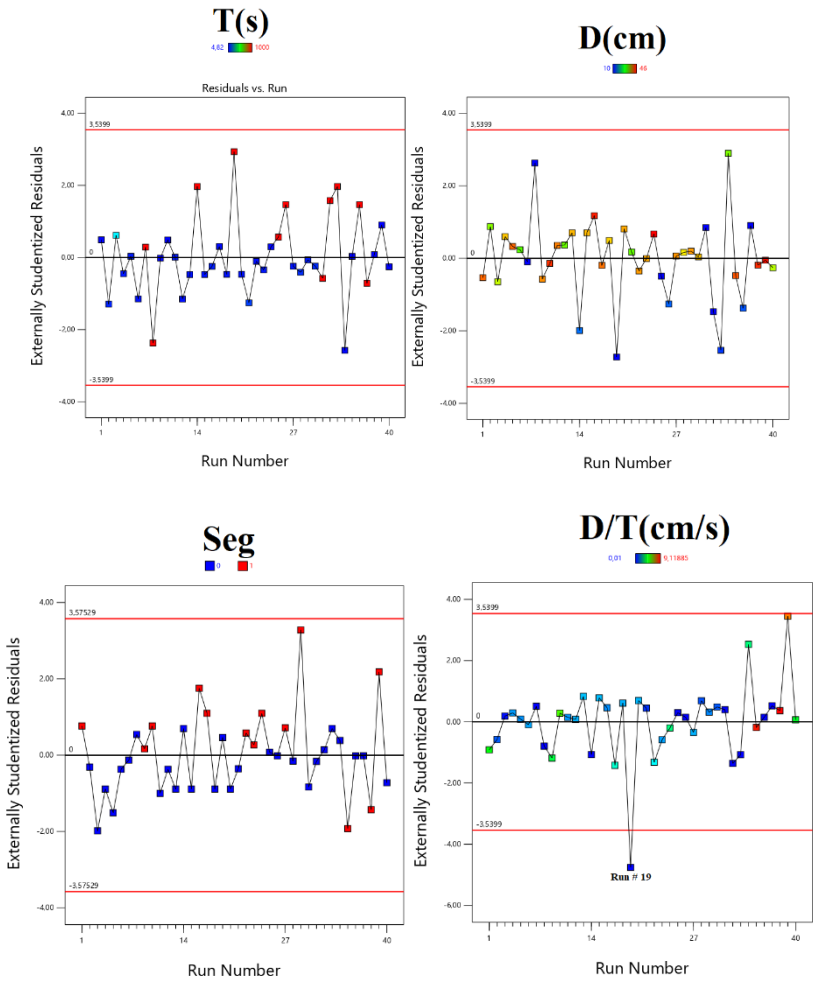


Figure 3: Residuals vs. Run. *T*(s): flow time. *D*(cm): spread of paste. Seg: segregation of the pastes. *D*/*T*(cm/s): Flow speed.

According to Figure 2, it can be seen that the normal distribution is acceptable. Figure 3 shows good residuals values as a function of the experiments except for the result of experiment 19 concerning the *D/T* (cm/s) response. This obtained value could be explained by the absence of SP in the paste making it less fluid, thus hindering the measurement of T and amplifying the error of the *D/T* (cm/s) response. In addition, this paste contains a high proportion of GP compared to similar pastes (e.g., Run 34) with the highest Water/Cement+GP ratio (equal to 1) for this category of pastes where SP = 0.

ANOVA was used to determine the statistical significance of the terms in the fitted models. Non-significant terms are removed from the model, which is noted as the "modified model". Table 6 presents the estimated coefficients of the significant terms for each response. It should be noticed that the segregation model (Seg) is not significant.

Table 6: Estimated Coefficients, ANOVA of models *T*(s), *D*(cm), Seg and *D/T* (cm/s) and factors percentages contribution (only significant models and factors are presented)

Models	**Factors**	**Estimated Coefficient**	**ANOVA**					**% of contribution**
				SS	***Df***	**MS**	***F* value**	**Probability *p***	
T(s)	C- SP	-82311.37		9.870E+05	1	9.870E+05	19.34	0.0001***	13.34
	GP- SP	-82399.34		1.007E+06	1	1.007E+06	19.73	< 0.0001***	13.10
	SP-W	-73655.18		8.119E+05	1	8.119E+05	15.91	0.0003***	10.76
			Res.	1.684E+06	33	51039.02			
			Tot. (co.)	7.541E+06	39				
D(cm)	C-SP	2207.85		710.16	1	710.16	29.44	< 0.0001***	10.59
	GP- SP	2223.26		733.13	1	733.13	30.39	< 0.0001***	10.25
	SP- W	2071.37		642.14	1	642.14	26.62	< 0.0001***	9.27
			Res.	796.03	33	24.12			
			Tot. (co.)	6920.22	39				
D/T(cm/s)	C- W	-14.82		9.34	1	9.34	22.15	< 0.0001***	4.22
	GP- W	-17.89		14.47	1	14.47	34.29	< 0.0001***	6.53
	SP- W	66.42		18.98	1	18.98	45.00	< 0.0001***	8.57
			Res.	13.92	33	0.4219			
			Tot. (co.)	221.42	39				

T(s): flow time, *D*(cm): spread diameter, Seg: segregation, *D*/*T*(cm/s): flow speed, *SS:* Sums of squares, *Df*: degrees of freedom, *S* : mean squares; Res: residual; Tot. (co.): total (corrected). The differences were considered significant at $p = 0.01$ to 0.05 (*), highly significant at $p = 0.001$ to 0.01(**) and very highly significant at $p \leq 0.001$ (***)

To determine the order of effectiveness of input factors, the absolute values of estimated coefficients in each tested response are examined (Table 6). The higher the absolute value of the coefficient, the more effective the factor. The order of effectiveness of input factors for each tested response, based on the absolute values of estimated coefficients, is as follows: GP-SP followed by C-SP, and finally SP-W for the response *T*(s); GP-SP followed by C-SP, and finally P-W for the response *D*(cm); SP-W followed by GP-W, and finally C-W for the response *D/T* (cm/s).

The percentages of contribution indicate the proportion of total variation in the response that can be attributed to each factor, which can be calculated using the sums of squares (SS) for each factor relative to the total sum of squares (Tot. (co.)) in each tested response. The percentages of contribution for each factor are reported in Table 6.

From Table 7, it can be seen that R^2 values are improved after modification of the models. The predicted R^2 values are in reasonable agreement with the *adjusted* R^2 since their difference is less than 0.2. The *predicted* R^2 which measures the predictive ability of the model shows a clear improvement for the modified models. The *Adeq Precision* measures the signal to noise ratio. Thus, a ratio higher than 4 is desirable. The ratio is 16.7 for T, 21.98 for *D* and 34.34 for *D/T* which indicates an adequate signal. All fit statistics improved with the modification of the models (Table 7).

Table 7: Fit Statistics for full and modified model.

Responses	**Model**	**Fit Statistics**			
		R²	***Adjusted R²***	***Predicted R²***	***Adeq Precision***
T(s)	Full	0.80	0.70	0.19	10.33
	Modified*	0.78	0.74	0.63	16.72
D(cm)	Full	0.90	0.85	0.58	14.30
	Modified*	0.89	0.86	0.81	21.98
D/T(cm/s)	Full	0.96	0.94	0.86	29.08
	Modified*	0.94	0.93	0.88	34.34

* The insignificant model terms are removed.

The modified models can therefore be used to explore experimental space which can be presented as ternary curves.

3.3 Effects of Constituents on Responses

3.3.1 Effects of Constituents on *T* (s)

Cement-SP, GP-SP and SP-Water interactions are the most significant terms on the variation of the T(s) response. The superplasticizer (SP) has therefore an important influence on the flow time of the pastes. This predominant role of SP on the flow of mixtures has already been observed [28]. Thus, the increase of SP in the mix reduces the flow time resulting in a more fluid paste, thus a gain in workability (Figure 4). These results are in agreement with other studies [29, 30].

The effects of cement and GP are almost identical (Figure 4). Indeed, fines in a mixture lead to a loss of workability due to the increase of the specific surface area of the mixture which requires more water to achieve the same workability. Cement and GP have generally the same negative effect on the rheology of mixtures and require more water or the use of a deflocculating agent such as SP to achieve the desired workability [14, 31]. Furthermore, SP and water play the same role, and both produce workable materials.

The interaction of SP with cement and GP highlights the importance of its role in fines deflocculating, promoting their movement in the mixture, thus allowing the expected rheological performance to be achieved. Studies from the literature have underlined the important effect of GP, specifically, on the fluidity of mixes. This effect, which may be either favourable or the opposite, is closely related to the type of SP used. Indeed, it has been reported that the fluidity decreases with increasing GP content in the presence of polycarboxylate type

SP similar to the one used in the present case. The opposite occurs in the case of the use of PNS type SP [31]. In addition to providing possible explanations for the existing opinion differences, these results confirm the strong interaction between GP and SP.
It has been noticed that SP has little influence on *T*(s) when the interaction of SP with water is beyond 0.02 (Figure 5). Indeed, superplasticizer is generally characterized by an optimum content called saturation dosage, beyond which their increase has no influence on the fluidity of the mixtures [32, 31].
The positive effect of water is not very pronounced, as shown in Figure 5, due to the restricted range in which this component varies.

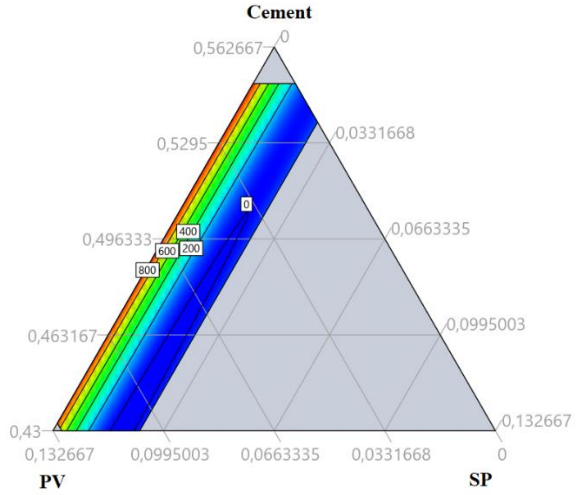


Figure 4: Ternary response diagram *T*(s) at Water = 0.473

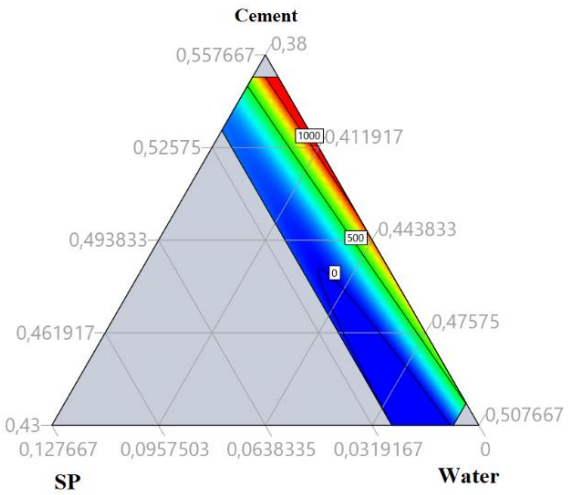


Figure 5: Ternary response diagram *T*(s) at GP = 0.062

3.3.2 Effect of Constituents on *D*(cm)

The significant interactions effects on the variation of the *D*(cm) response are: Cement-SP, GP-SP and GP-Water where SP remains the most influential constituent (Table 6). According to Figure 6, increasing the percentage of SP in the mix leads to an increase in *D*(cm). The same is true for the proportion of water in the mix (Figure 7). Cement and GP have the same influence on the paste spread, but it is less important than the influence of SP. Unlike the latter, which has a positive effect on workability, cement and GP produce a negative effect. Increasing their dosage leads to a drop in workability [33]. Overall, the spreading results are in agreement with the results of *T* where SP positively influences the flowability [31].
The addition of GP to the mix causes the spread diameter to be reduced and thus decreases the workability. This loss of workability has been attributed to the increase in fines in the mix which increases the cohesion of the mix and hinders their flow [33].
Solimane and Tagnit-Hamou [34] also noted the loss of fluidity with the incorporation of GP. They attributed this loss to the lower density of GP (2500 kg/m^3) than that of cement (3060 kg/m^3). With the partial replacement of cement by GP, the volume ratio of solid particles to water becomes larger with an increasing substitution rate. Thus, the friction between the solid particles in the paste increases.

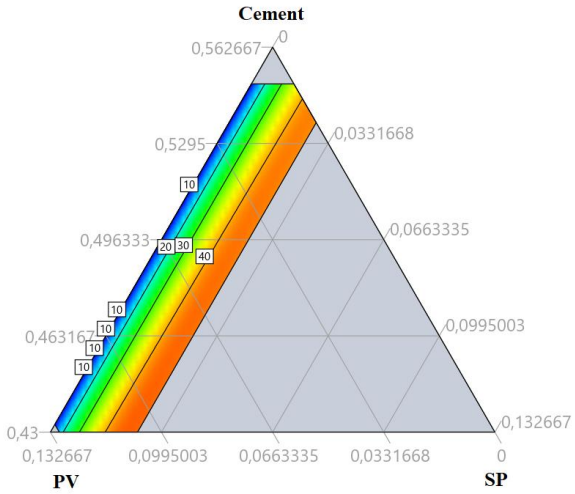


Figure 6: Ternary response diagram *D*(cm) at water = 0.473

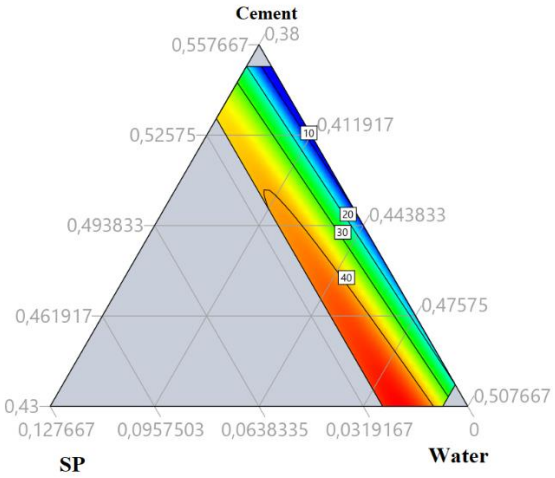


Figure 7: Ternary response diagram *D*(cm) at GP = 0.062

3.3.3 Effects of Constituents on *D/T*(cm/s)

The interest of this response lies in the fact that it combines *T*(s) and *D*(cm) into a single response that reflects the flow velocity. The latter is high for the more workable pastes.
The influential terms differ from those observed for *T* and *D*. Cement-Water, GP-Water and SP-Water interactions are the most significant terms highlighting the importance of water on the paste flow velocity (Table 6). Figure 8 shows clearly that *D/T* increases with increasing water content in the paste.

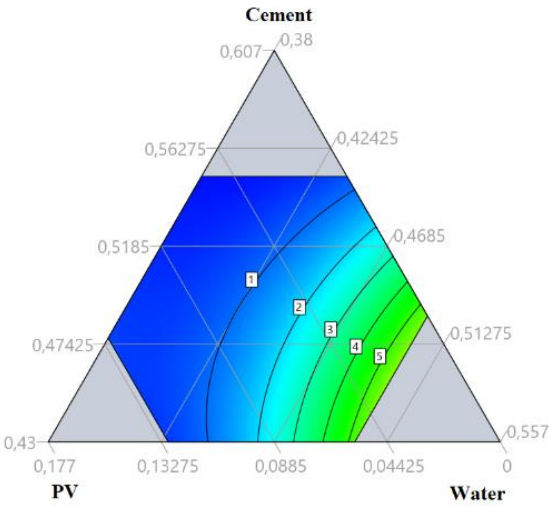


Figure 8: Ternary response diagram *D/T*(cm/s) at SP = 0.013

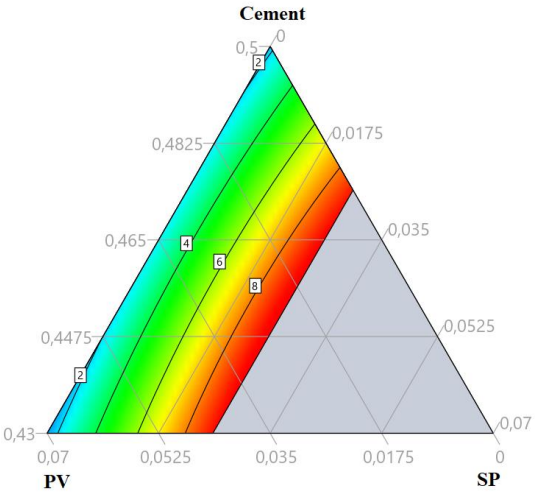


Figure 9: Ternary response diagram *D/T* (cm/s) at water = 0.5

The variation of the *D/T*(cm/s) response also depends on the proportion of SP in the paste. Indeed, when it increases the *D/T* response increases too (Figure 9).

The ratio of the *D*(cm) to *T*(s) response is relevant as a response because it shows the importance of the proportion of water on the workability of the paste which has not been shown by analyzing independently each response. It should also be noted that the *D/T*(cm/s) ratio model has better fit statistics (R^2, R^2_{adj}, R^2_{pre}, *Adeq Precision*) compared to the *T*(s) and *D*(cm) models (Table 6). This new variable, not yet considered in current research, is a promising one and could constitute a reference parameter for self-placing pastes and even fluid concretes, such as the water/cement ratio for cementitious mixes in general. Further studies to examine this parameter are recommended.

3.4 Optimization and Model Validation

Multi-objective optimization results in several optimal solutions of different desirability. Solutions with high desirability indicate that the objective can be easily achieved. For this purpose, two optimal solutions with a desirability value of 0.84 are selected for validation. The optimal compositions and predicted responses of the SCP1 and SCP2 mixtures are summarized in Table 8.

Table 8: Optimal composition of the two pastes minimizing *T*, maximizing *D* and *D/T* for Desirability = 0.845

	C	**GP**	**SP**	**W**
SCP1	0.430	0.130	0.018	0.422
SCP2	0.452	0.039	0.008	0.500

The predictive accuracy of the model has been examined by conducting additional experiments under optimal conditions and comparing the predicted values to the measured values obtained with the SCP1 and SCP2 pastes. For each point, the experiment was repeated 3 times.
The measured and predicted results are grouped in Table 9. The t-test shows that the measured results are not statistically different from the predicted values. The predicted optimal solutions are therefore validated.

Table 9: Multi-response optimization validation responses

	Responses	**Predicted Response** μ	**Experimental Mean** m	$\lvert t \rvert *$	**signification**
SCP1	*T* (s)	39.88	36.32 ± 3.53	1.75	N.S
	D (cm)	40.08	38.92 ± 1.04	1.94	N.S
	D/*T* (cm/s)	1.01	1.08 ± 0.13	1.67	N.S
SCP2	*T* (s)	7.69	7.71 ± 0.41	0.07	N.S
	D (cm)	37.81	39.56 ± 2.08	1.46	N.S
	D/*T* (cm/s)	4.87	5.14 ± 0.47	1.73	N.S

3.5 Mechanical Strength of Optimal Pastes

The comparison of the mechanical strength results of the two optimal pastes as reported in Table 10, allows to note that the performances obtained at 28 days for SCP1, containing 25 % of partial substitution of cement by GP, are lower than those obtained for SCP2 which contains

only 7 % GP for cement substitution. Indeed, the flexural tensile strength of SCP1 is -5.4% lower than that of SCP2. As for the compressive strength, it goes from 37.60 MPa for SCP2 to 32.50 MPa for SCP1, i.e. a reduction of -13.56 %. This drop in mechanical performance is explained by the reduction in the quantity of cement which is replaced by GP. Therefore, SCP1, with a higher substitution rate, develops lower strengths at 28 days.

Table 10. Mechanical strength of optimal pastes

	Flexion (MPa)		**Compression (MPa)**	
	28 days	**90 days**	**28 days**	**90 days**
SCP1 (25%GP)	3.30	4.60	32.50	43.87
SCP2 (7 % GP)	3.49	4.40	37.60	37.92
Difference %	-5.40	+4.50	-13.56	+15.69

On the other hand, a reversal of this trend is observed at 90 days where the strength values are higher for SCP1. Indeed, the flexural tensile strength increases from 4.40 MPa for SCP2 to 4.60 MPa for SCP1 with a gain of +4.5%. As for the compressive strength, it increases from 37.92 MPa for SCP2 to 43.87 MPa for SCP1, which represents an increase of +15.69%. Overall, at 90 days, the strengths are higher in the case of the highest GP substitution rate, especially for SCP1 which contains more GP at a rate of 25% of cement replacement.
These results are very interesting and allow to retain the single optimal formula SCP1. Also, they show the long-term effectiveness of GP, because although the performances were reduced at 28 days because of the reduction of the quantity of cement compared to the elevated GP content, then the tendency was completely reversed at 90 days with significant gains in strength. This increase in mechanical performance is justified by the pozzolanic effect of GP due to the reaction of its silica with the lime released from the hydration of cement [35]. This reaction is similar to that of cement to form hydrated calcium silicates, C-S-H; but is slower and only occurs in the long term [36]. Several studies have obtained similar results where the focus has been on the GP pozzolanic activity which improves cement hydration. Its addition to cementitious mixtures by partial substitution of cement has been successfully used by achieving long-term strength gains compared to 28-day strengths [37, 36].

4 Conclusion

A mixture design was applied to investigate the workability of GP-based pastes as a function of the proportions of cement, GP, SP and water.
The models for the variation of *T*(s) and *D*(cm) are statistically significant and highlight the importance of the SP-GP interaction where the SP would act on the fines deflocculating of the GP to achieve the expected rheological performance.
The *D/T* (cm/s) ratio is a new approach to analyze the results of *T*(s) and *D*(cm) by combining the two. The model reflecting the variation of this response has the best fit statistics. The *D/T*(cm/s) response allowed to highlight the effect of water contrary to the analysis carried out on each response separately despite its important role in the rheology of mixtures. This response requires further work to make it a reference in the study of self-compacting mixtures as is the water/cement ratio for the mechanical performance of concretes, pastes, and mortars.

The proportions for two optimal pastes have been predicted: SCP1 and SCP2 containing 7 and 25% GP respectively. The validation of the optimal solutions by experiments shows the predictive accuracy of the models fitted for flow time *T*(s), spreading diameter *D*(cm) and flow velocity *D/T*(cm/s).

Evaluation of the flexural tensile and compressive strengths of the optimal pastes showed the positive effect of GP on long-term mechanical performance. SCP1 with 25% GP has higher strengths at 90 days than SCP2 with 7% cement replacement by GP. In addition, it offers an economic gain because the quantity of cement in the mix is reduced, thus leading to an ecological gain by reducing the emission of CO_2 gas due to the manufacture of cement. The pozzolanic character of GP has allowed the long-term mechanical performance to be achieved and even exceeded. By comparing the strengths of SCP2 with a lower substitution GP content and therefore with more cement, it has been noticed that the loss of mechanical performance due to the high substitution rate is largely recovered with time.

The present work allowed the determination of the optimal GP dosage of 25% as partial substitution of cement, leading thus to a self-compacting paste with the best mechanical performance.

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