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Optimization of mix proportions of mineral aggregates using Box Behnken design of experiments

M. Muthukumar, D. Mohan *, M. Rajendran

Department of Chemical Engineering, A.C. College of Technology, Anna University, Chennai 600 025, India

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Abstract

Optimization of mix proportions of silica aggregates for use in polymer concrete was attempted using statistical techniques. High purity silica aggregates of six different standard particle sizes were chosen for the study. Void content of 54 statistically designed combinations were experimentally determined by adopting standard technique. Using Design Expert software the results were analyzed and an optimum composition having minimum void content was achieved. The optimum combination had a correlation coefficient of 0.95782 which proved the fitness of the selected model in analyzing the experimental data.

Keywords: Aggregate gradation; Design expert; Optimization; Polymer concrete; Regression analysis; Statistical techniques; Void

1. Introduction

Polymer concrete is a composite material in which the binder consists entirely of a synthetic organic polymer, and where the aggregates are bound together by the polymer matrix. It is also commonly known as synthetic resin concrete, plastic resin concrete or simply resin concrete as per ACI Committee 548 [1]. The aggregates used in polymer concrete are graded silica, quartz, crushed stone, gravel, limestone, chalk, condensed silica fume (silica flour, silica dust), granite, clay, expanded glass, metallic fillers etc. The distribution of aggregates should be such as to allow for a minimum void volume for dry packed aggregates which will result in dense packing. This minimizes the amount of binder required to assure proper bonding of all the aggregate particles. Normally the binder content ranges from 5% to 15% of the total weight but if the aggregate mix is fine, it may even require up to 30% binder.

Dense packing of aggregates in the polymer concrete matrix results in better properties. To achieve this, we can either minimize the void content of the aggregate mix where the binder requirement will be less or use loosely packed aggregate mix with higher binder content. A low binder requirement for filling the voids between the particles in the mixture reduces shrinkage and other related properties of polymer concrete systems. Also the cost of the composition will be lesser thereby making it more economically attractive.

Aggregate mix proportions for use in polymer concrete were reviewed. Most of the works like those of Ehrenburg [2], Golterman et al. [3], Popovics [4] in optimizing aggregate combination for minimizing voids have been for making high strength portland cement concrete where the binder is cement and the aggregate sizes are much larger than those used for polymer concretes. Though the importance of minimum void content of aggregate mixes for use in polymer concrete has long been realized, the literature [5,6] contains very few references in this regard. Ohama [5], Kantha Rao and Krishnamoorthy [6] have studied the effects of aggregate mix proportions based on practical considerations. In the present study the aggregate mix proportions of mineral aggregate having minimum void content suitable for polymer concretes were proposed based on statistical techniques.

* Corresponding author. Fax: +91-44-235-0240.
E-mail addresses: dmohan@annauniv.edu, mohantarun@yahoo.com (D. Mohan).

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2. Design of experiments

The classical approach of changing one variable at a time and studying the effect of the variable on the response is a complicated technique, particularly in a multivariate system or if more than one response are of importance. Design of experiments are statistical techniques which can be used for optimizing such multivariable systems. Using design of experiments based on response surface methodology, the aggregate mix proportions having least void content can be arrived with minimum number of experiments without the need for studying all possible combinations experimentally. Further the input levels of the different variables for a particular level of response can also be determined.

Response surface methodology comprises a group of statistical techniques for model building and model exploitation. By careful design and analysis of experiments, it seeks to relate a response or output variable to the levels of a number of predictors or input variables that affect it. It allows calculations to be made of the response at intermediate levels which were not experimentally studied and shows the direction in which to move if we wish to change the input levels so as to decrease or increase the response [7–9]. Response surface methodology using Box Behnken [10] design was used to optimize the response of six input variables. The optimization process involves studying the response of the statistically designed combinations, estimating the coefficients by fitting it in a mathematical model that fits best the experimental conditions, predicting the response of the fitted model and checking the adequacy of the model.

The six variables chosen for the study are designated as A, B, C, D, E, F and the predicted response, namely void content, is designated as Y. Details of particle size, lower limit, upper limit of the input variables are given in Table 1. The mathematical relationship between the six variables and the response can be approximated by the second order polynomial:

$$\begin{aligned} Y = {} & \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_4 x_4 + \beta_5 x_5 + \beta_6 x_6 \\ & + \beta_{11} x_1^2 + \beta_{22} x_2^2 + \beta_{33} x_3^2 + \beta_{44} x_4^2 + \beta_{55} x_5^2 + \beta_{66} x_6^2 \\ & + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 + \beta_{14} x_1 x_4 + \beta_{15} x_1 x_5 + \beta_{16} x_1 x_6 \\ & + \beta_{23} x_2 x_3 + \beta_{24} x_2 x_4 + \beta_{25} x_2 x_5 + \beta_{26} x_2 x_6 + \beta_{34} x_3 x_4 \\ & + \beta_{35} x_3 x_5 + \beta_{36} x_3 x_6 + \beta_{45} x_4 x_5 + \beta_{46} x_4 x_6 + \beta_{56} x_5 x_6 \end{aligned}$$

where β_0 is a constant; β_1, β_2, β_3, β_4, β_5, β_6 are linear coefficients; β_{12}, β_{13}, β_{14}, β_{15}, β_{16}, β_{23}, β_{24}, β_{25}, β_{26}, β_{34}, β_{35}, β_{36}, β_{45}, β_{46}, β_{56} are cross product coefficients; β_{11}, β_{22}, β_{33}, β_{44}, β_{55}, β_{66} are quadratic coefficients; and $x_1 = (A - x_0)/\Delta x$, x_1 = coded value of the variable A, x_0 = value of A at the center point, Δx = step change; $x_2 = (B - x_0)/\Delta x$ and so on where A, B etc. are the input variables.

Table 1
Levels of variables chosen for the design

S. no	Grade	Particle size, mm		Input level, g (coded value)		
		Min	Max	Low	Medium	High
1	A	4.76	9.52	0 (−1)	50 (0)	100 (+1)
2	B	2.38	4.76	0 (−1)	50 (0)	100 (+1)
3	C	1.19	2.38	0 (−1)	50 (0)	100 (+1)
4	D	0.60	1.19	0 (−1)	50 (0)	100 (+1)
5	E	0.30	0.60	0 (−1)	50 (0)	100 (+1)
6	F	0.15	0.30	0 (−1)	50 (0)	100 (+1)

3. Materials and methods

Naturally occurring silica sand from mines were washed, dried and sieved into six different grades, varying in sizes from 0.15 to 9.72 mm as indicated in Table 1. The silica particles chosen for the study are high purity, high silica content, iron free and very low acid soluble type suitable for use in chemical resistance application.

Bulk densities were determined for the 54 different combinations by adopting standard techniques [11]. Using the bulk density, void content was determined using the formula:

$$\%\mathrm{VC} = \frac{(\mathrm{SG} - \mathrm{BD}) \times 100}{\mathrm{SG}}$$

where VC = void content, SG = specific gravity of the aggregate, BD = Bulk density of the aggregate mix.

4. Results and discussion

The present investigation was carried out to minimize the void content to a practically lower level and optimize the mix proportion of the individual variables. Void content (response) for the 54 statistically designed combinations (inputs) suggested by Box Behnken design of experiments for the six variables were experimentally determined and the minimum and maximum void content obtained were 21.96% and 31.4% respectively. Using Design Expert software (Statease Inc., Minneapolis, MN 55103) the experimentally obtained void content were analyzed and the coefficients of the parameters in the second order polynomial were arrived at and given by the regression equation (which gives the predicted void content) shown below regardless of their significance:

$$\begin{aligned}\text{Predicted void content} = {} & 24.810 - (1.986x_1) \\ & - (1.147x_2) - (0.132x_3) \\ & + (1.210x_4) + (1.088x_5) \\ & - (0.520x_6) + (0.602x_1^2) \\ & + (0.571x_2^2) + (0.239x_3^2) \\ & + (0.192x_4^2) + (0.684x_5^2) \\ & + (1.401x_6^2) + (1.155x_1x_2) \\ & + (0.443x_1x_3) + (0.032x_1x_4) \\ & - (0.791x_1x_5) - (0.120x_1x_6) \\ & + (0.899x_2x_3) + (0.030x_2x_4) \\ & - (0.713x_2x_5) - (0.609x_2x_6) \\ & - (0.375x_3x_4) - (0.981x_3x_5) \\ & - (0.939x_3x_6) - (0.281x_4x_5) \\ & - (0.580x_4x_6) + (1.336x_5x_6)\end{aligned}$$

The above equation is based on the quadratic model suggested by the software against other models such as linear, cubic and 2 factor interactions (2FI), since this quadratic model fitted well with the experimental data. The fitness of the above model with respect to minimizing void content can be explained from the

Table 2
Lack of fit test

Source	Lack of fit			F value
	Sum of squares	DF	Mean square	
Linear	130.2947	42	3.10225	18.75607
2FI	54.82327	27	2.03049	12.27625
Quadratic	26.23894	21	1.24947	7.55425
Cubic	1.51271	3	0.50423	3.04858

Pure error sum of squares: 0.827.
Pure error DF: 5.
Pure error mean square: 0.16540.

Table 3
Model summary statistics

Source	Standard deviation	R squared	R value
Linear	1.670276	0.599971	0.774578
2FI	1.318738	0.830221	0.911165
Quadratic	1.020293	0.917427	0.957824
Cubic	0.540799	0.992862	0.996425

Table 4
ANOVA for response surface quadratic model

Source	Sum of squares	DF	Mean square	F value	Prob > F
Model	300.71	27	11.138	10.699	<0.0001
x_1	94.68	1	94.685	90.956	<0.0001
x_2	31.56	1	31.556	30.313	<0.0001
x_3	0.42	1	0.419	0.402	0.5315
x_4	35.11	1	35.114	33.731	<0.0001
x_5	28.41	1	28.406	27.287	<0.0001
x_6	6.48	1	6.479	6.224	0.0193
x_1^2	3.73	1	3.730	3.583	0.0695
x_2^2	3.35	1	3.348	3.216	0.0845
x_3^2	0.59	1	0.587	0.564	0.4594
x_4^2	0.38	1	0.380	0.365	0.5509
x_5^2	4.82	1	4.817	4.627	0.0409
x_6^2	20.20	1	20.200	19.404	0.0002
x_1x_2	10.67	1	10.672	10.252	0.0036
x_1x_3	1.57	1	1.566	1.505	0.2309
x_1x_4	0.02	1	0.016	0.016	0.9015
x_1x_5	5.01	1	5.009	4.811	0.0374
x_1x_6	0.12	1	0.115	0.111	0.7421
x_2x_3	6.46	1	6.462	6.208	0.0194
x_2x_4	0.01	1	0.007	0.007	0.9344
x_2x_5	8.12	1	8.123	7.803	0.0097
x_2x_6	2.96	1	2.965	2.848	0.1035
x_3x_4	1.13	1	1.125	1.081	0.3081
x_3x_5	7.70	1	7.703	7.399	0.0115
x_3x_6	14.10	1	14.100	13.545	0.0011
x_4x_5	0.63	1	0.633	0.608	0.4426
x_4x_6	2.69	1	2.691	2.585	0.1199
x_5x_6	14.28	1	14.285	13.722	0.0010
Residual	27.07	26	1.041		
Lack of fit	26.24	21	1.249	7.554	0.0169
Pure error	0.83	5	0.165		
Cor. total	327.78	53			

lack of fit test given in Table 2, model summary statistics given in Table 3 and analysis of variance (ANOVA) given in Table 4. The different statistical terms used here along with their explanations are listed in Appendix A.

Lack of fit test compares the residual error to the pure error from replication and gives F values for all the models. F value must be lower if a particular model is to be significant. From the F test, it was found that the calculated F value was more than the tabulated F value

Table 5
Box Behnken design for the six variables and their experimental and predicted response (%void content)

Run	*A*	*B*	*C*	*D*	*E*	*F*	Experimental response	Predicted response
1	−1	−1	0	−1	0	0	30.16	29.32
2	0	−1	−1	0	−1	0	25.46	25.70
3	0	1	0	0	1	1	26.37	26.90
4	0	0	−1	1	0	1	27.83	28.20
5	0	0	−1	−1	0	1	27.62	26.19
6	1	0	0	1	−1	0	24.96	25.53
7	−1	−1	0	1	0	0	31.28	31.61
8	−1	0	1	0	0	1	27.37	27.13
9	1	0	0	1	1	0	26.12	25.56
10	1	0	−1	0	0	1	24.87	25.06
11	0	0	1	−1	0	−1	27.25	26.55
12	−1	0	−1	0	0	−1	29.41	29.07
13	0	−1	0	0	1	−1	29.53	28.99
14	−1	0	0	1	−1	0	28.08	27.85
15	1	−1	0	−1	0	0	21.96	22.97
16	1	0	0	−1	−1	0	23.83	22.48
17	−1	0	−1	0	0	1	30.66	30.15
18	0	0	1	1	0	−1	28.28	29.38
19	0	−1	1	0	1	0	27.91	27.24
20	0	−1	0	0	−1	−1	28.28	28.06
21	1	0	1	0	0	1	23.79	23.80
22	−1	0	0	1	1	0	30.12	31.05
23	0	1	1	0	1	0	25.87	25.32
24	−1	1	0	1	0	0	27.66	27.07
25	0	−1	0	0	1	1	30.28	31.84
26	0	1	0	0	−1	−1	30.28	28.41
27	0	0	0	0	0	0	25.25	24.81
28	−1	0	0	−1	−1	0	23.96	24.94
29	1	1	0	−1	0	0	22.84	22.93
30	0	0	0	0	0	0	24.33	24.81
31	0	–	0	0	−1	1	25.62	25.57
32	0	1	0	0	−1	1	23.25	23.48
33	0	−1	1	0	−1	0	25.33	25.60
34	−1	0	0	−1	1	0	29.41	29.26
35	0	0	1	−1	0	1	24.25	24.80
36	0	1	1	0	−1	0	26.08	26.53
37	0	0	0	0	0	0	24.83	24.81
38	1	0	0	−1	1	0	23.83	23.64
39	0	0	0	0	0	0	25.33	24.81
40	0	0	1	1	0	1	26.50	25.31
41	0	1	−1	0	−1	0	22.05	23.03
42	−1	1	0	−1	0	0	24.13	24.65
43	0	−1	−1	0	1	0	31.40	31.26
44	0	0	−1	1	0	−1	28.74	28.52
45	−1	0	1	0	0	−1	29.66	29.80
46	1	0	−1	0	0	−1	24.54	24.46
47	0	0	−1	−1	0	−1	22.67	24.19
48	0	1	−1	0	1	0	26.33	25.75
49	1	0	1	0	0	−1	26.12	26.96
50	0	1	0	0	1	−1	26.12	26.49
51	0	0	0	0	0	0	24.50	24.81
52	1	−1	0	1	0	0	26.33	25.39
53	0	0	0	0	0	0	24.62	24.81
54	1	1	0	1	0	0	25.04	25.47

Table 6
Experimental combination vs optimized combination

S. no	Variable	Experimental combination having minimum void			Optimized combination having minimum void		
		Coded	Uncoded	%	Coded	Uncoded	%
1	*A*	1	100	40	0.81	90.5	39.6
2	*B*	−1	0	0	0.53	76.5	33.5
3	*C*	0	50	20	−1.0	0	0
4	*D*	−1	0	0	−1.0	0	0
5	*E*	0	50	20	−1.0	0	0
6	*F*	0	50	20	0.23	61.5	26.9
	%void		21.96			21.002	

for the corresponding degrees of freedom thus rejecting the null hypothesis of "significant lack of fit" for linear, 2FI and quadratic models indicating that the lack of fit is insignificant. Quadratic being the higher order polynomial was selected among them. For the cubic model, the calculated F value was less than the tabulated F value for corresponding degrees of freedom, thus accepting the null hypothesis, indicating that the lack of fit is significant thereby making this model aliased and hence was disqualified.

Model summary statistics gives several comparative measures for model selection. Ignoring the aliased model, the quadratic model comes out the best: low standard deviation and high R squared statistics.

ANOVA for response surface quadratic model gives the sum of squares and degrees of freedom for the model terms from which mean square of the model terms are calculated. F value of the quadratic model and individual model terms helps in finding their significance. The model F value of 10.70 implies that the model is significant. Values of "Prob > F" less than 0.0500 indicate significant model terms. Thus x_1, x_2, x_4, x_5, x_6, x_5^2, x_6^2, x_1x_2, x_1x_5, x_2x_3, x_2x_5, x_3x_5, x_3x_6, x_5x_6 are significant model terms. Among the different models studied, only

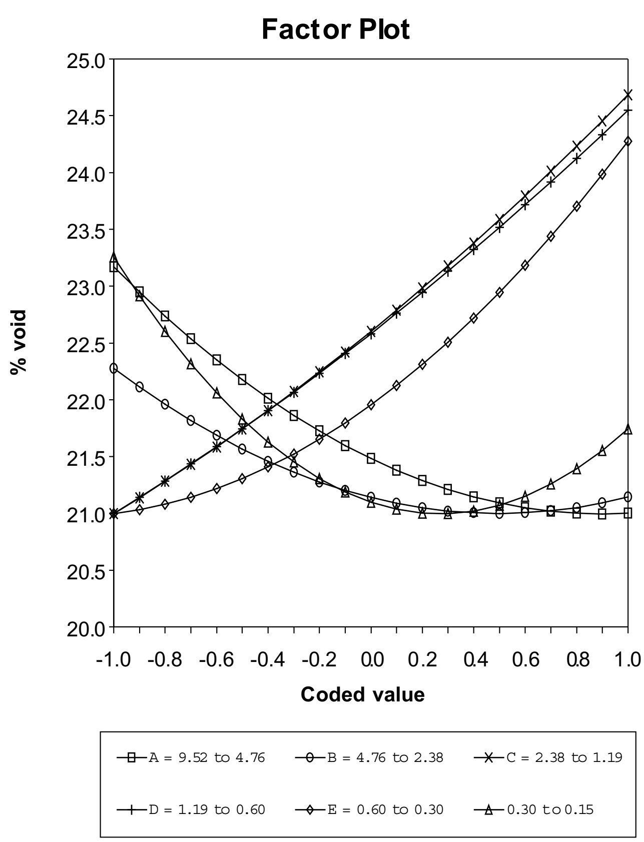

Fig. 1. Factor plot.

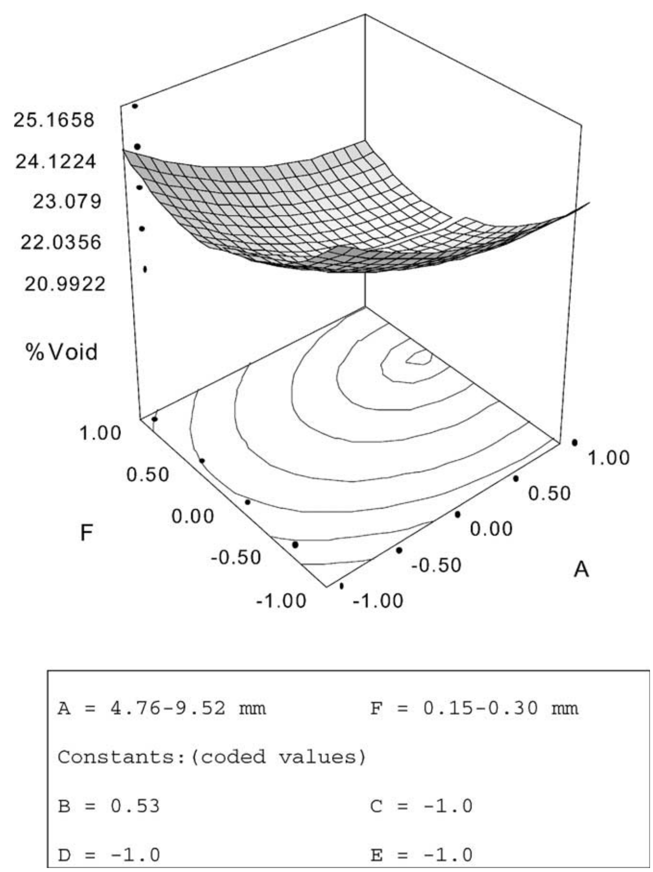

Fig. 2. 3D diagram showing the contour plots for grades *A* and *F* on the void content.

the quadratic model was found to fit the experimental data best with a correlation coefficient (R value) of 0.95782 which is closer to unity, the ideal condition, indicating that the fitness of the selected model is good and the model could be used for further navigation [12].

The experimentally determined void content along with the predicted void content obtained from the regression equation for the 54 combinations are given in Table 5. By optimization, the minimum void content given by the software was 21.002% corresponding to coded values of variables $A = 0.81$; $B = 0.53$; $C = -1.00$; $D = -1.00$; $E = -1.00$; $F = 0.23$ and are compared in Table 6 with the experimentally obtained minimum void content.

The optimized combination showed lesser void content than the experimentally studied combinations and was also different from them. This proves the usefulness of statistical techniques in model exploitation and empirical model building.

5. Effect of variables

The effect of individual variables on the void content of the mix, when all the other variables are kept at their optimum level, is shown in the factor plot Fig. 1. All individual variables were found to have their own effect on the void content.

As the proportion of grade A in the mix increases from -1 to $+1$ in coded values, the void content decreases. This shows that grade A contributes significantly in reducing the void content. Grade A being the largest among the sizes chosen, higher input value will result in lesser requirement for other grades to fill the inter particle voids. Similarly, as the proportion of grade B in the mix increases from -1 to $+1$ in coded values,the void content decreases up to 0.51 corresponding to the optimum value and thereafter starts increasing. As the proportion of grades C, D and E in the mix increases from -1 to $+1$ (individually) in coded values, the void content increases. This shows that grades C, D and E do not contribute in minimizing the void content. As the proportion of grade F increases from -1 to $+1$ in coded values, the void decreases up to 0.23 corresponding to the optimum value and thereafter increases. The 3D

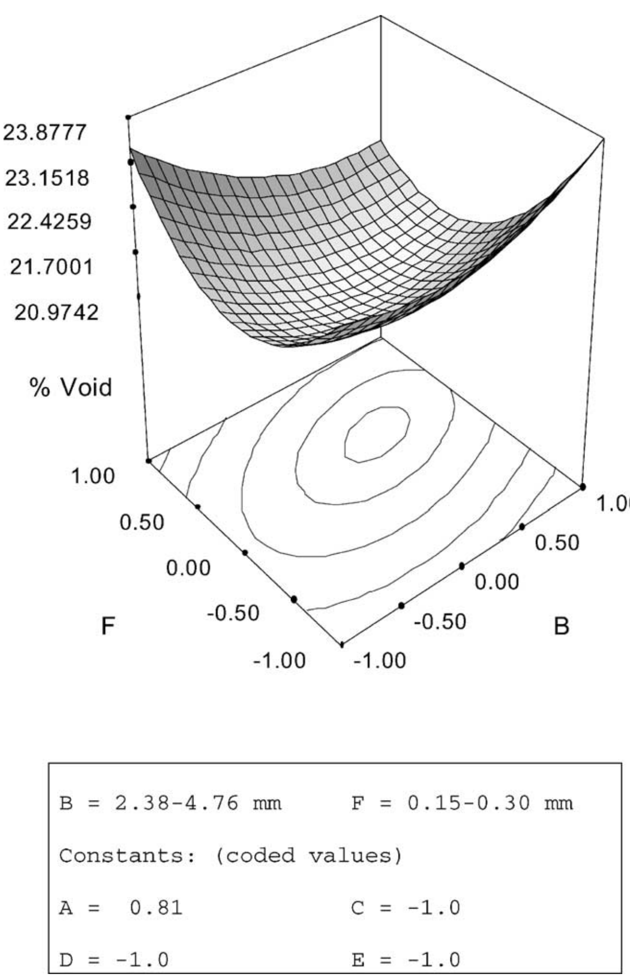

Fig. 3. 3D diagram showing the contour plots for grades B and F on the void content.

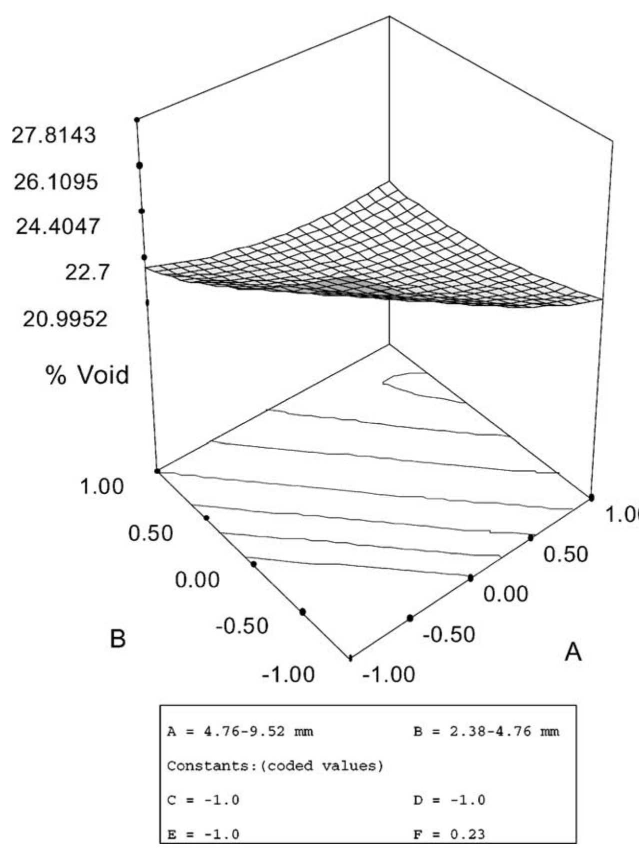

Fig. 4. 3D diagram showing the contour plots for grades A and B on the void content.

contour plots of the effect of three significant variables A, B and F on the void content are shown in Figs. 2–4.

6. Conclusion

Void content for different sizes of silica particles was optimized using Box Behnken design of experiments on the basis of regression analysis of experimental data. It was concluded that, from limited number of experimental runs and with the help of Design Expert software, that out of the six different particle sizes chosen for the study, only three of them were found to be sufficient for obtaining a mix with minimum void content, thereby eliminating the other three grades. Thus a gap graded aggregate composition was obtained which had fine particles filling the inter particle voids between the coarser aggregates. The optimum combination had a void content less than the experimentally determined void content with a correlation coefficient of 0.95782 which proved the fitness of the selected model in analyzing the experimental data.

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The author wishes to thank Mr Mark Anderson of Statease Inc., Minneapolis, MN 55103 for his suggestions and advise on using the Design Expert software.

Appendix A

Statistical terms and their definitions

Model sum of squares	Total of the sum of squares for the terms in the model
Residual sum of squares	Total of the sum of squares of all the terms not included in the model
Lack of fit sum of squares	Residual sum of squares after removing the pure error sum of squares
Pure error sum of squares	Sum of squares from replicated points
Model DF	Number of model terms including intercept −1
Residual DF	Corrected total DF minus the model DF
Lack of fit DF	Amount of information available after accounting for blocking model terms and pure error
Pure error DF	Amount of information available from replicated points
Mean square	Sum of squares divided by DF
Model F value	A test for comparing model variance with residual variance. If the variances are close to the same the ratio will be close to one and it is less likely that any of the factors have a significant effect on the response calculated by model mean square divided by residual mean square
Term F value	Test for comparing term variance with residual variance. If the variances are close to the same, the ratio will be close to 1 and it is less likely that the term has a significant effect on the response. Calculated by term mean square divided by residual mean square
Lack of fit F value	Test for comparing lack of fit variance with pure error variance. If the variances are close to the same, the ratio will be close to 1 and it is less likely that lack of fit is significant
Prob $> F$	Probability of seeing the observed F value if the null hypothesis is true. Small probability values call for rejection of the null hypothesis. The probability equals the proportion of the area under the curve of the F-distribution that lies beyond the observed F value. The F distribution itself is determined by the degrees of freedom associated with the variances being compared
R squared	A measure of the amount of deviation around the mean explained by the model. $1 - (\text{SS residual}/(\text{SS model} + \text{SS residual}))$
Adj. R squared	A measure of the amount of variation around the mean explained by the model adjusted for the number of terms in the model. The adjusted R squared decreases as the number of terms in the model increases if those additional terms don't add value to the model. $1-((\text{SS residual}/\text{DF residual})/((\text{SS model}+\text{SS residual})/(\text{DF model}+\text{DF residual})))$
Pure error	Amount of variation in the response in replicated design points
Cor. total	Totals of all information corrected for the mean

Coefficient of variation	Standard deviation expressed as a % of mean, calculated by dividing the standard deviation by the mean and multiplying by 100
Residual	Difference between experimental and predicted points. Consists of terms used to estimate experimental error

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