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Remediation of heavy metal contaminated soils by solidification/stabilization with fly ash, quick lime and blast furnace slag

Ramiz Raja* and Supriya Pal

Department of Civil Engineering, National Institute of Technology Durgapur, Durgapur-713 209, West Bengal, India

E-mail: ramiz.nitdgp@gmail.com

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This paper presents a study on the stabilization of contaminated soil deposits using fly ash (FA), quick lime (QL) and blast furnace slag (BFS). The soil samples were collected from a landfill site at Arupota, near EM Bypass, Dhapa, Kolkata. The soil was found polluted with heavy metals viz. Zn, Pb, Cu and Cr. The maximum dry density (MDD) and unsoaked California Bearing Ratio (CBR) values of the soil were found 1320 kg/m^3 and 4.82% respectively. An attempt was made to immobilize the contaminants as well as improve the characteristic strength in the soil simultaneously through mechanical mixing with stabilizers. The factor-based Box-Behnken design (BBD) of Response Surface Methodology (RSM) was applied to examine the combined effect of selected additives that is, FA (5–20%), QL (5–20%) and BFS (5–20%), on soil MDD and CBR values which were considered as response functions. The predicted values of MDD and CBR in the model were found to be in close agreement with experimental values (R^2 = 0.8265 and 0.9371 for MDD and CBR respectively). The optimum values of FA, QL and BFS were found to be 5%, 20% and 20% respectively, with a desirability of 0.942. Under these conditions, the MDD and CBR values were found to be 1450 kg/m^3 and 28.60% respectively. The improved characteristics due to the combined application of additives in the soil render suitability of the RSM method in quantifying the stabilizer dosage.

Keywords: Contaminated soil, immobilization, stabilization, optimization, strength parameters.

Introduction

The geo-environmental engineers nowadays are facing great challenges to abate the soil pollution levels in urban and semi-urban localities which happened due to indiscriminate and unscientific disposal of solid and liquid wastes. Conventionally, the top soils in these sites are removed/replaced by a good quality soils which is expensive, time-consuming and sometimes impracticable due to nonavail-ability of large quantities of soils. On the other hand, soil stabilization with additives is a good alternate solution to immobilize the contaminants and improvement of strength properties of soils simultaneously. The reason behind immobilization phenomenon was due to the stabilized soil possesses lower fraction exchangeable content and higher fraction of residual content than the untreated soil[1].

This study focuses to use fly ash (FA), quick lime (QL) and blast furnace slag (BFS) easily available local ingredients as stabilizer for the improvement of various engineering properties of contaminated soil. Most of the earlier studies also used similar or different stabilizers for improvement of engineering properties of soil. Sharma *et al.* studied the combined effect of lime and fly ash on strength properties of clayey soils[2]. Zhang *et al.* studied stabilization treatment of contaminated soil: a field scale application in Shanghai[3].

Objective:

The present paper deals with the novelty of using locally available sustainable and inexpensive materials such as FA, QL and BFS collectively to examine their interaction effect on strength gaining process of soil. As works on stabilization of contaminated soil are not well documented in the literature, it is expected that the outcome of the present endeavour will be helpful for geo-environmental engineers to study more effectively the problems concerned, and thereby reclaiming the contaminated land.

Materials and methods:

Study area:

The municipal solid waste (MSW) landfill site popularly known as Dhapa at Arupota near Eastern Metropolitan (EM) bypass in the eastern fringe of the metropolis of Kolkata, India, has been selected for the study area in this research.

A vast area of Dhapa, adjoining the current core MSW dump site had been evolving from the raw MSW dumping. It is currently used for the cultivation of vegetables for the adjoining localities.

Collection of soil sample:

The soil samples were collected from the study area by auger boring from a depth of 1 m below the ground surface and preserved in polythene bags and transported to the Soil Mechanics and Foundation Engineering Laboratory of Civil Engineering Department, National Institute of Technology Durgapur. The samples were collected from three different points at 10 m apart.

Tests for physical properties of soil samples:

The collected soil samples were first oven dried at 60 ±2°C for 24 h in the laboratory and then the dry soil was powdered by wooden mallet for performing the following physical test as per the standard procedures depicted in Bureau of Indian Standard codes[4] specified for particle size distribution, specific gravity, Atterberg limits, light proctor compaction, optimum moisture content as depicted in the soil characteristics section subsequently.

Box-Behnken experimental design:

A design of matrix as shown in Table 1 were prepared taking the three additive components showing ranges as input parameters in Box-Behnken experimental design in response surface methodology (RSM). The Design Expert software 8.0.6.1 (trial version, Stat-Ease, Minneapolis, Minnesota, USA) has been used for the purpose.

In this study, a three-level, three-factor Box-Behnken experimental design was applied to explore the effects of three independent variables, namely, FA, QL and BFS on MDD and CBR value. The low, middle and high levels of each factor were coded as –1, 0 and +1, respectively. The coded and actual values of the three input factors are shown in Table 2.

Essentially, the Box-Behnken experimental design is a spherical, revolving design consisting of the central and middle points of the edges of a cube circumscribed on a sphere[5]. However, it can also be considered as consisting of three interlocking 2^2 factorial designs and a central point[6]. The Box-Behnken design requires an experimental number according to $N = n^2 + n + c_p$, where n is the factor number and c_p is the replicate number of the central point[7]. This methodology is often applied to optimise the waste treatment process[8,9]. It is claimed to be more efficient compared to other response surface models such as central composite, D-optimal and full factorial designs. This method examines the ef-

Table 1. Box-Behnken experimental design for the three independent variables used in the study together with observed and predicted response

Std.	Run	FA (%)	Lime (%)	BFS (%)	Observed MDD (kg/m^3)	Observed CBR (%)	Predicted MDD (kg/m^3)	Predicted CBR (%)
1	1	5	5	12.5	1332	19.86	1323.8225	16.1075
17	2	12.5	12.5	12.5	1367	12.3	1363.68125	9.62
15	3	12.5	12.5	12.5	1362	11.68	1363.68125	9.62
6	4	20	12.5	5	1350	11.99	1319	8.27
11	5	12.5	5	20	1410	17.66	1374.86	15.7325
3	6	5	20	12.5	1398	21.83	1388.015	17.2325
9	7	12.5	5	5	1320	20	1329.935	20.3825
4	8	20	20	12.5	1346	11.37	1361.915	9.8825
5	9	5	12.5	5	1310	22.743	1303.475	23.495
2	10	20	5	12.5	1364	12.3	1380.9725	11.0075
12	11	12.5	20	20	1490	30.31	1457.39	23.6075
7	12	5	12.5	20	1370	19.09	1408.3625	17.72
10	13	12.5	20	5	1280	15.37	1292.54	12.5075
16	14	12.5	12.5	12.5	1365	11.99	1363.68125	9.62
14	15	12.5	12.5	12.5	1354	11.68	1363.68125	9.62
8	16	20	12.5	20	1410	26.98	1423.8875	20.495
13	17	12.5	12.5	12.5	1369	12.3	1363.68125	9.62

Table 2. Independent variables and their levels used for response surface design

Variables (%)	Symbols	Range and levels		
		Low (–1)	Middle (0)	High (+1)
FA	A	5	12.5	20
QL	B	5	12.5	20
BFS	C	5	12.5	20

fects of individual factors, as well as higher-order interactions with limited number of experimental runs[10,11]. In this case, 17 experiments were performed, with five repeat experiments at the design centre to check the variance in the experimental data and for the error estimation. The experimental results of Box-Behnken design experiments are shown in Table 1. Linear and second-order polynomial models were developed using multiple regression analysis to study the interactive effect of the influencing factors following Box-Behnken models. The sequential F test, lack-of-fit test and other adequacy measures were carried out in selecting the best-fit model[12]. Considering all linear, square and linear-to-linear interactions, the model equation for predicting the optimal point is given below expression:

$$Y = \beta_0 + \sum_{i=1}^{n} \beta_i x_i + \sum_{i=1}^{n} \beta_{ii} x_i^2 + \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} \beta_{ij} x_i x_j + \varepsilon \qquad (1)$$

where, Y is the response (dependent variable); β_0 is the constant coefficient; β_i is the linear coefficient; β_{ii} is the quadratic coefficient; β_{ij} is the interaction coefficient; $x_i x_j$ are factors (independent variables); and ε is the residual error. The optimum values of the selected variables were evaluated by solving the regression equation and by response surface contour plot analysis following the method described by Montgomery[10]. The range of dependent variables was explained by the multiple coefficient of determination, R^2. The model equation has also been applied to predict the optimum value and, subsequently, to explicate the interaction between factors within the specified range following by Elibol and Ozer[13].

Results and discussion

Soil characteristics:

The physical properties of the soil are shown in Table 3. The percentage of fines (silt and clay) of the soil is 56.75%. The liquid limit and plastic limit of soil cannot be determined as the soil is non-plastic in nature.

Table 3. Collected soil properties

Property	Experimental result	Standard method
Specific gravity	2.31	IS-2720 - Part 3, 1980
Optimum moisture content	23.52	IS-2720 - Part 7, 1997
Dry density (kg/m^3)	1320	IS-2720 - Part 7, 1997
CBR value (%) unsoaked	4.82	IS-2720 - Part 16, 1987
pH value	7.64	IS-2720 - Part 26, 1987
Organic Matter (%)	4.68	Loss on ignition method

Heavy metal characteristics:

Soils of the test sites are extremely contaminated with heavy metals. The landfill soils had the significantly higher amount of lead, zinc, copper compared to those in the background soil. Thus the soil is also responsible for the Surface Water and Ground Water pollution of Dhapa through heavy metal contamination. The heavy-metal concentrations are also shown in Table 4. The permissible limit of the heavy-metals as per Canadian Soil Quality Guidelines are also given in the table due to the lack of well-defined standard in India (CCME, 2007)[14].

Table 4. Heavy metal concentration of the collected soil

Heavy metal	Standard method	Average result	Permissible value	Unit
Chromium (Cr)	USEPA3052/3051A	28.93	64	mg/kg
Lead (Pb)	USEPA3052/3051A	335.04	70	mg/kg
Zinc (Zn)	USEPA3052/3051A	564.28	200	mg/kg
Copper (Cu)	USEPA3052/3051A	193.96	63	mg/kg

Box-Behnken experimental design:

The results of each experiment based on the Box-Behnken experimental design are also shown in Table 1. Four response functions, namely, linear, two-factor interaction, quadratic and cubic, were selected to develop a regression eq. (1) which correlated the experimental data of MDD and CBR value. The sequential model sum-of square tests, lack-of-fit tests and model summary statistics were also performed to examine the adequacy of the aforementioned models. The cubic model was found to be aliased. The sequential model sum of squares and model summary statistics showed that the quadratic model was found to be the best fit to the experimental data in both cases of MDD and CBR values. In the case of MDD the lowest p-value (< 0.0001), standard deviation (0.025) and the predicted R^2 (0.8265) were found.

In the case of CBR value the lowest *p*-value (< 0.0001), standard deviation (2.25) and the predicted R^2 (0.9371) were found. The empirical relationship between the response and the input variables can be expressed by the following quadratic model regression equation (in coded terms):

$$Y_1 = 1.27 + 0.00566\ A - 0.000533\ B + 0.00033\ C - 0.00037\ AB - 8.63\times10^{19}\ AC + 0.000533\ BC$$

$$Y_2 = 62.12 - 2.04\ A - 1.75\ B - 4.41\ C - 0.01\ AB + 0.08\ AC + 0.07\ BC + 0.03\ A^2 + 0.04\ B^2 + 0.11\ C^2$$

where, Y_1 is MDD, A is FA, B is QL, C is BFS, Y_2 is CBR value.

Effect of various factors on MDD and CBR value:

Contour plots were drawn to examine the combined effect of any two factors of the response, while another factor was set at the middle level using the Box-Behnken experimental design. The MDD and CBR value of the soil is affected by both FA and BFS dosage as shown in Figs. 1 and 4. The maximum MDD and CBR value was achieved at FA content of 5% and at BFS content of 20% while other factor remained at the middle level. Further increase of FA content did not produce any significant improvement in MDD value. The MDD and CBR value of the soil is affected also by both FA and QL dosage as shown in Figs. 2 and 5. The maximum MDD and CBR value was achieved at FA content of 5% and at QL content of 20% while other factor remained at the middle level. The combined effect of QL and BFS dosage on MDD and CBR value of the soil are plotted in Figs. 3 and 6. The maximum MDD and CBR value was achieved at QL content

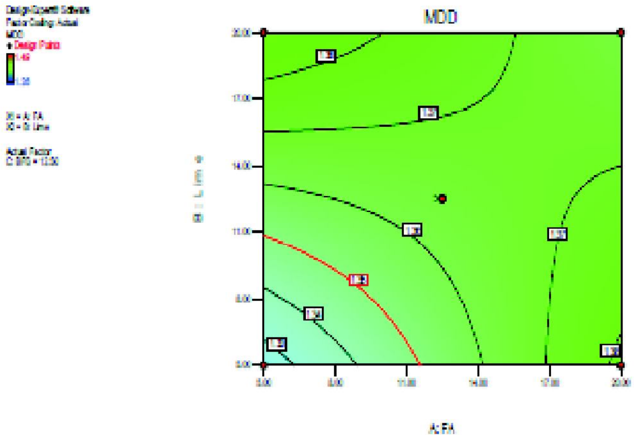

Fig. 2. Contour plot for MDD of the soil is affected by both FA and lime dosage.

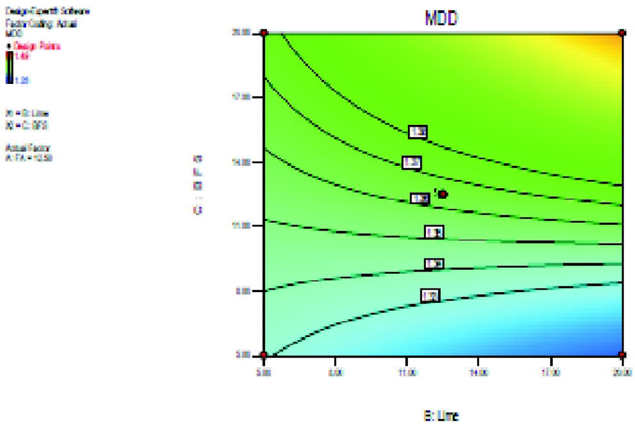

Fig. 3. Contour plot for MDD of the soil is affected by both lime and BFS dosage.

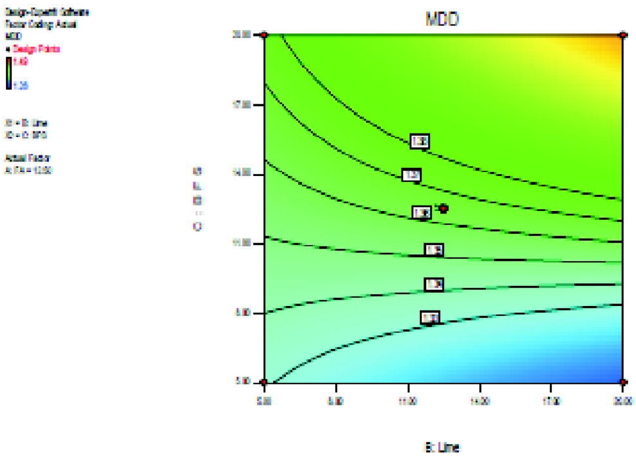

Fig. 1. Contour plot for MDD of the soil is affected by both FA and BFS dosage.

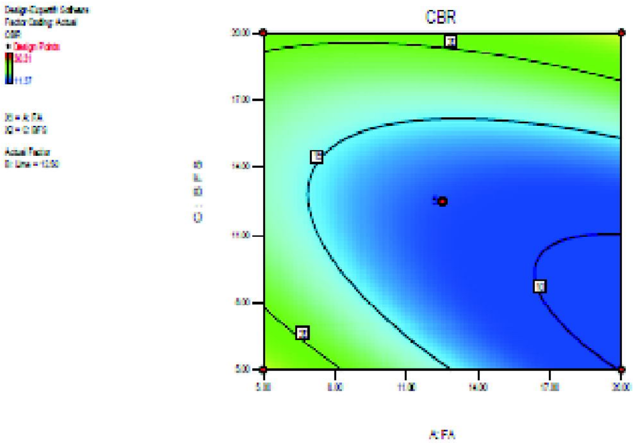

Fig. 4. Contour plot for CBR of the soil is affected by both FA and BFS dosage.

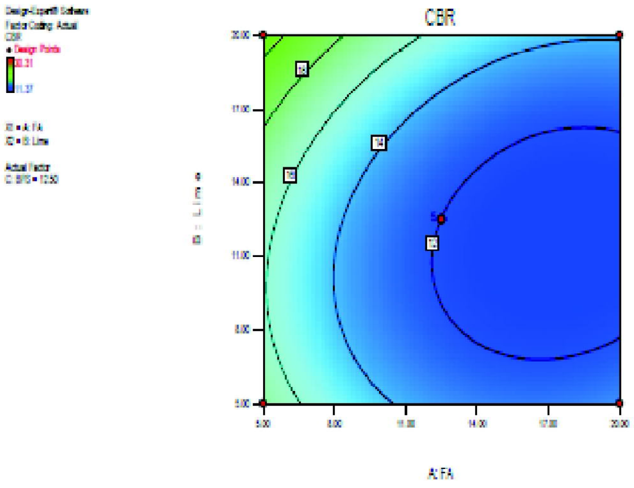

Fig. 5. Contour plot for CBR of the soil is affected by both FA and lime dosage.

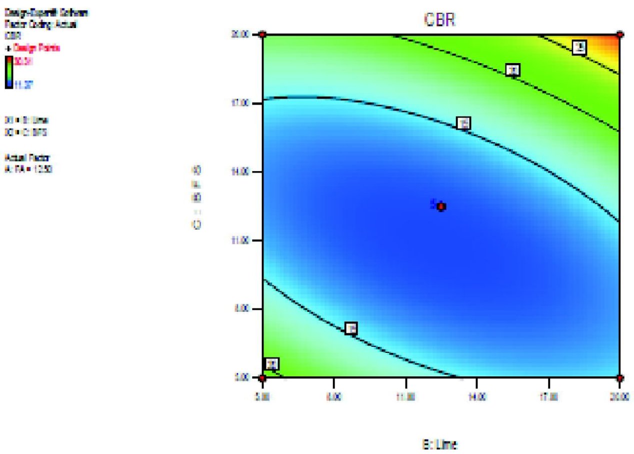

Fig. 6. Contour plot for CBR of the soil is affected by both lime and FS dosage.

of 20% and at BFS content of 20% while other factor remained at a constant level.

Optimisation using the desirability function:

In the present research, a desirability function was also applied in order to estimate the optimum input experimental data to maximise the MDD and CBR values of soil along with minimisation of the additive dosage[15]. The numerical optimisation determines a point that maximises the desirability function by searching in the design space[16]. The desired goal for each factor is first fixed. The goals have the following options: maximum, minimum, target and within the range. The minimum and maximum value of each included parameters should be provided. A relative weight is also assigned to each goal in order to adjust the shape of the particular desirability function. The goals are then combined to an overall desirability function. Desirability is an objective function that varies between 0 (outside the limits) and 1 (at the goal). The function starts from a random point and moves to a steep slope to a maximum. There may be several local maxima in the curvature of the response surface and their combination into desirability function. To find the global maximum, the function is started from several random points. The multiple response method was examined to optimise any combination of three goals, namely, FA (%), QL (%) and BFS (%). With the use of a desirability function with a prefixed goal and weight for each factor, the global maximum was found at FA at 5%, QL at 20% and BFS at 20% with a desirability of 0.942. The desirability ramp plot (Fig. 7) shows the optimal experimental conditions for the improvement of MDD and CBR values at maximum desirability. Additional confirmatory experiment was conducted in the laboratory using optimal values of FA, QL and BFS. The MDD and CBR value were found 1450 kg/m^3 and 28.60%. This result is in reasonable agreement with experimental results and optimisation analysis indicates the suitability of the developed quadratic model, and it may also be noted that these optimal values are valid within the specified range of process parameters as considered in the present study. The Box-Behnken experimental design in conjunction with the desirability function can be effectively used for the optimisation of additive constant for reclamation of contaminated land by increasing strength properties.

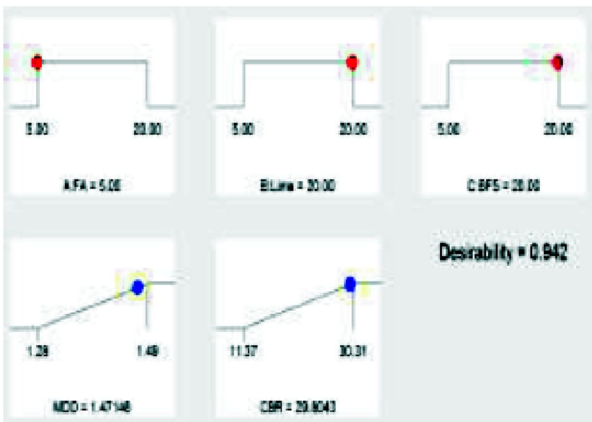

Fig. 7. Desirability ramp plot for numerical optimisation.

Conclusions

An attempt has been made in the present study to use the mixture of FA and QL and BFS as stabilizers to enhance strength parameters of the contaminated soil collected from a MSW landfill site. The following specific conclusions can be made on basis of the test results obtained:

(i) The fly ash, quick lime and blast furnace slags are found strong stabilizing candidates for improving the strength properties of heavy metal contaminated soil through mechanical mixing.

(ii) The BBD model in RSM with the application of desirability function can be used as effective tools in optimizing the stabilizers to achieve maximum gain in characteristic strength of the contaminated soil.

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