DOE and ANN Models for Powder Mixture Packing

Design of experiments and artificial neural network techniques were used to study packing of fused alumina powders composed of three different sizes of particles.

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Packing of powder compacts always has been of interest to ceramists because of the potential benefits of a well-packed greenware. When greenware is densely packed, the final sintered ware is denser and has improved firing shrinkage, bulk density and mechanical strength properties. Therefore, ceramics must be formed using well-packed mixtures of particles. Significant work has been accomplished on packing of powder mixtures. A review of such work has been done by Funk and Dinger.[1]

Generally, in the ceramics field, systems are constituted of continuous size distribution of irregular particles, and random packing can be obtained. Packing can be further subdivided into two types: random dense and random loose. In random dense packing, the particles have been agitated or pressed to attain the closest packing possible without long-range order, which is equivalent to the tapped bulk density. In random loose packing, the lowest stable packing density without introducing long-range order or deformation is attained, which is equivalent to aerated bulk density.[2]

It has been shown that the efficiency of classical models is quite good when the morphology of particles approximates a sphere. However, the models fail when the particles have irregular shapes.[3] Little theoretical or experimental work can be found in the literature concerning the packing of irregular particles.[4] Some attempts have been made to describe the deviation of particle shape from spherical.[5]

Particle shape and container shape and dimension have an effect on particle packing. It has been shown that, with a decrease of sphericity, the porosity of a randomly packed bed of uniform-sized particles decreases.[6] Empirical relations between packing efficiency and particle size have been proposed by Ayer and Soppet.[7] Karlsson and Spring[8] have presented a modification of the Furnas equation.

The dimension of the container with respect to particles size is another important parameter. Leva and Grummer[9] have studied the packing of nonspherical particles in cylinders. They have shown that porosity increases with an increase of the particle-diameter:container-diameter ratio. However, little work has been done on modeling the packing of particles, except for a few studies that involve statistical techniques[10] and artificial neural networks (ANNs).[11]

Guerin[11] has worked on the prediction of commercial alumina powder compact density using ANNs, but the powders used in the study were not fused and, hence, not fully dense. Another study on packing prediction has been done by Silva et al.[10] They have used statistical experimental design techniques to create a response surface of the particle compact densities.

Romagnoli and Rivasi[12] have shown the possibility of using the design of experiments (DOE) approach in the packing optimization of nonspherical spray-dried powders used in the production of ceramic tile by pressing. However, powder compact densities of fully dense fused particles have not yet been studied using ANN

or DOE. Comparison of the two techniques for their effectiveness in predicting the packing of powder compacts appears to be interesting. ANNs are best suited for modeling large sets of data, and the DOE technique is best suited for a small number of experiments.

In this study, two techniques have been used for prediction of powder compact porosities in mixtures of three different-sized fused alumina powders. The first is the mixture design technique that produces a polynomial model of the powder-packing system. The second is the ANN technique that is extensively used to model complex systems in many fields.[13,14]

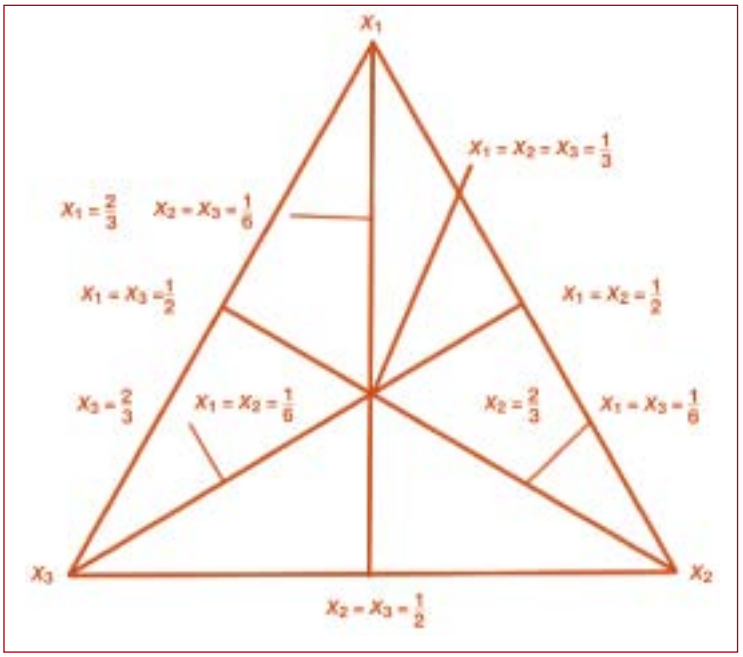

Fig. 1 Mixtures that were used to create the DOE model (x_1 is coarse, x_2 is medium and x_3 is fine).

The methodological approach used is mixture design. This approach is different from the more widely used "trial and error" approach. The statistical method of mixture design, a part of DOE, can be used to study the influences of two or more additives. It is a structured and organized method for determining the relationship between the components and the output of that process. Correct experimental planning provides more information with less effort and decreases the subjectivity of the results, which increases technical and scientific values. Correct experimental planning generates a map of the response over a specified region of formulation. It is possible to discover the critical variables, define mathematical models and, using them, optimize the product and the industrial process.[15,16]

Data Collection

Three sizes (3, 30 and 350 μm) of fused alumina powder were mixed and uniaxially compacted in the form of cylindrical pellets to measure their packing ability in the green state. Porosities of the cylindrical pellets were generated by preparing powder mixtures in such proportions that were planned using mixture design, which is a DOE method.

Fifty-six sets of data were generated and used for two separate studies. The first study was the DOE method, which produced a polynomial model with r^2 = 0.9202. The second study involved ANNs that required random partition of the data set into two sets to be used for model creation and model validation of the ANN models. A multilayer feed-forward backpropagation (MFFB) learning algorithm was used as an ANN tool to predict porosity, which was the response variable. Based on the training data, an ANN model of porosity as a function of constituent mix proportions was created with low average error levels (2.7%). Testing of the model also was performed successfully, with r^2 = 0.8913. The results indicated that the mixture model and the ANN model provided good predictions for powder packing.

Table 1 Composition and Porosity of Augmented Simplex-Lattice Design Blends

Standard order	Run order	Coarse (wt%)	Medium (wt%)	Fine (wt%)	Experimental porosity (%)
1	10	100.00	0.00	0.00	34.2
2	5	0.00	100.00	0.00	44.8
3	3	0.00	0.00	100.00	45.9
4	13	50.00	50.00	0.00	32.9
5	1	50.00	0.00	50.00	32.8
6	7	0.00	50.00	50.00	35.8
7	6	66.67	16.67	16.67	27.2
8	4	16.67	66.67	16.67	34.3
9	8	16.67	16.67	66.67	39.2
10	11	0.00	50.00	50.00	36.0
11	12	100.00	0.00	0.00	35.1
12	9	0.00	0.00	100.00	45.9
13	14	0.00	100.00	0.00	44.8
14	2	33.33	33.33	33.33	30.3

The three sizes of fused alumina powders were obtained from Treibacher Co., Austria.[17] Powder particle sizes were narrowly distributed,

because these powders were obtained from an abrasive manufacturer that must meet strict limitations for sizes (FEPA).[18] Various blends (15 g each) were made from the powders using an augmented simplex-lattice design (Fig. 1). Four samples were replicated to estimate pure error and to test lack-of-fit (LOF).

This estimate of pure error came from replication of several design points equal to the number of components plus one, up to a maximum of five. Each interval was constituted by a continuous size distribution. The run order for experiments was randomized to counteract any time-related effects. The analysis of the results was conducted using design-expert 6.0.10 (Stat-Easy Inc.).

The powder specimen preparation procedure was as follows. Distilled water (9 g) was added to the powder mixture (15 g). The suspension was stirred using a magnetic stirrer for 2 h. The agglomerates were oven dried at 110°C for 2 h and broken using a mortar and pestle. Twenty drops of 5% poly(vinyl alcohol) solution and 3–5 mL of ethyl alcohol were added. The powder mixtures then were oven dried, homogenized using a mortar and pestle and pressed at 100 MPa in the form of cylindrical pellets of 15 mm diameter and 6–10 mm height. Height and diameter of pellets were measured using a precision caliper (Model CD-15CP, Mitutoyo), and porosities were calculated. More details about the procedure can be found in Ref. 19.

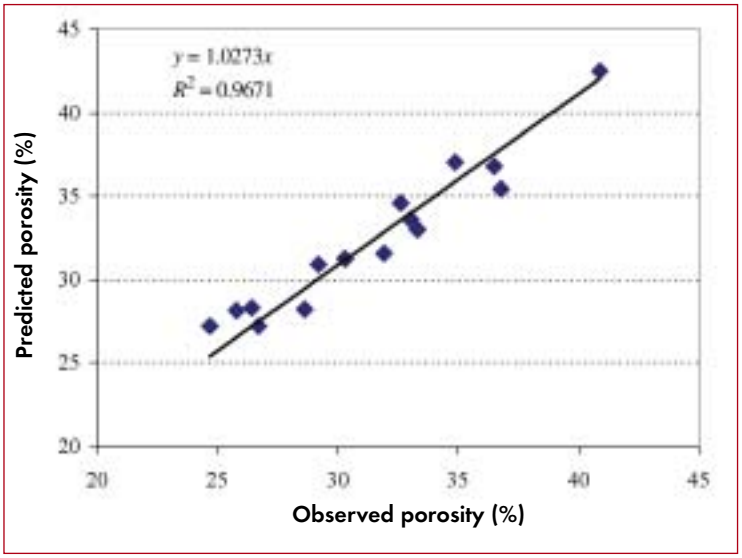

Fig. 2 Experimental versus calculated porosity for 15 samples. Calculated data were obtained using Eq. (1).

DOE Model

This first set of experiments used the DOE approach, which helped obtain a quadratic model for the mixtures. Data were generated from this 14-run, augmented simplex-lattice design (Table 1). An analysis of variance (ANOVA) table for this set of data also was prepared (Table 2). ANOVA analysis was used to remove insignificant terms. F-test and LOF confirmed the applicability of the quadratic model.

A mathematical model for blends has been determined using a response surface method:

$$P = 0.347C + 0.447M + 0.460F - 2.90 \times 10^{-3}CM - 3.07 \times 10^{-3}CF - 3.87 \times 10^{-3}MF - 6.46 \times 10^{-5}CF(C - F) \quad (1)$$

where P is porosity (in percent), C is fraction of coarse in the mixture (in percent), M is fraction of medium in the mixture (in percent) and F is fraction of fines in the mixture (in percent).

Table 2 Analysis of Variance for Mixture Design Experiments

Source†	Sum of squares	DF	Mean square	F value	Probability >F
Model	479.85	06	079.98	362.71	<0.0001
Linear mixture	188.76	02	094.38	428.03	<0.0001
AB	053.13	01	053.13	240.96	<0.0001
AC	060.93	01	060.93	276.36	<0.0001
BC	137.58	01	137.58	623.96	<0.0001
AC(A-C)	022.71	01	022.71	103.02	<0.0001
Residual	001.54	07	000.22		
LOF	001.12	03	000.37	003.51	0.1284
Pure error	000.42	04	000.11		
Corrected total	481.40	13			

†A is coarse, B is medium and C is fine size.

Coarse size distribution leads to lower porosity among the three size distributions considered. Equation (1) shows a lower first-order mixture-model coefficient (0.347) that predicts the response from the pure component. Medium and fine particles have similar effects on the porosity.

The second-order negative terms in Eq. (1) indicate synergism. In other words, they represent the positive effect of the blending on the decrease of porosity. Even if it is well-known in literature, the DOE approach shows numerically the size of such drop. The

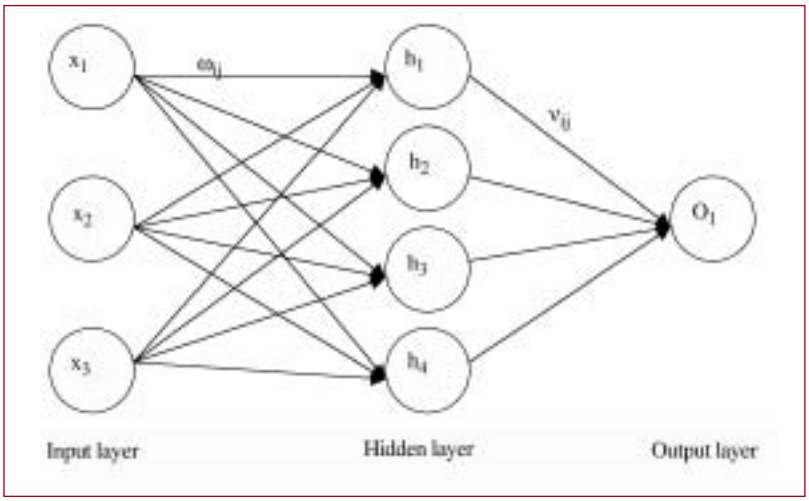

Fig. 3 ANN model architecture used in this study (x_i, h_i and o_i are input, hidden and output layer neurons; ω_{ij} and v_{ij} are weights for connections between layers of processing elements).

magnitudes of the exponents are two or four units less than first-order. However, they must be multiplied by the fractions (in percent) of two sizes (or three, as in the case of the last term of Eq. (1)) so that their contribution is not secondary. The effect of the fraction of medium and fine particles is the most important. However, the interaction of coarse–medium and coarse–fine sizes cannot be underestimated.

The mixture model fits the experimental data with a high grade of efficiency. The R^2 value is 0.9968, and the predicted R^2 of 0.9856 is in reasonable agreement with the adjusted R^2 of 0.9940. LOF, that is, the variation of the data around the fitted model, of 3.51 implies that it is not significant relative to the pure.

Fifteen more samples were prepared to check the efficiency of the model with respect to its ability to foresee the porosity of a blend when the size composition is new. Experimental versus calculated porosity results were compared (Fig. 2 and Table 3). The slope of the straight line was approximately one, and coefficient of correlation was high if the relevant number of samples was considered. In fact, the probability that theoretical and experimental data had an $r^2 \geq 0.9202$ without real correlation is <0.05%.

The mixture design approach permits optimization of size distribution to obtain a target value of porosity. On the basis of the model of Eq. (1), the best blend to have the lowest porosity is 71.91% coarse, 5.26% medium and 22.83% fine. Theoretically, this permits a porosity of 26.01%. The model can help to obtain the desired porosity. This possibility cannot be offered by other theoretical approaches, such as the Andreassen, Furnas or Funk–Dinger models. Moreover, it considers, via analysis of experimental results, the morphology of the particles and does not assume that they are spherical.

ANN Model

The computational details about ANN model construction are well documented in the literature.[14] Therefore, such information is not presented here. In this study, a three-layer feed-forward ANN architecture has been constructed. In the input layer, there are three neurons for the three input variables of the proportions of each powder (Fig. 3). In the hidden layer, four neurons have been chosen by trial and error. Finally, in the output layer, one neuron is used for the output variable of porosity.

The input variables are as follows:

- x_1 is the fraction of coarse (in percent) (powder code is F46);
- x_2 is the fraction of medium (in percent) (powder code is F320);
- x_3 is the fraction of fine (in percent) (powder code is F1200); and
- o is porosity (in percent).

Table 3 Powder Mixtures Used to Test DOE and ANN Models

Coarse (%)	Medium (%)	Fine (%)	Observed porosity (%)	ANN model predicted porosity (%)	DOE model predicted porosity (%)
20	70	10	36.8	36.8	35.4
50	30	20	26.4	27.3	28.3
10	50	40	32.6	33.7	34.6
20	60	20	33.0	33.8	33.6
70	30	0	31.9	30.4	31.6
30	40	30	30.3	30.2	31.2
10	10	80	40.9	40.8	42.5
10	30	60	36.5	35.0	36.8
60	10	30	26.7	27.7	27.2
40	20	40	29.2	29.3	30.9
60	20	20	24.7	27.1	27.2
80	10	10	25.8	29.3	28.1
70	20	10	28.6	28.1	28.2
30	60	10	33.3	33.4	33.0
0	70	30	34.9	35.8	37.0

Neurons in each layer were fully connected to every single neuron in the neighboring layers. No bias term was used during modeling, but a momentum term was used to help obtain faster convergence during the iterations. There were a total of 56 data sets, 41 of which were used for the training of the ANN. The remaining 15 were for testing of the model. Each data set had four components (x_1, x_2, x_3, o), three of which were the input variables and the fourth of which was the output variable of porosity.

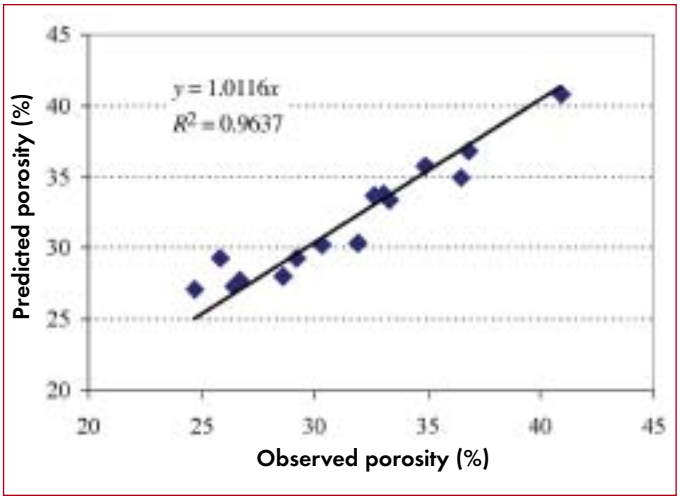

Fig. 4 Testing of the ANN model with 15 measured data.

The program operated for 80,000 iterations using the 41 data sets. The optimal weights were calculated with an average absolute error (PAAE) for learning of 2.7% for porosity (Table 4). The r^2 value for learning (training) was 0.9437. The model then was tested with physically measured data using the remaining 15 data sets, which also were used for testing the DOE model. The results of the testing runs (Table 3 and Fig. 4) showed a high magnitude of r^2 (0.8913), which indicated that the testing was successfully performed. Because of the limited number of experimental data, only 15 testing outputs were compared to real measured data. Large test data sets are always preferred when possible. The performance of the model was quite satisfactory (Table 3 and Fig. 4).

Sensitivity Analysis

Sensitivity analysis involves the use of the developed ANN model to predict outputs (porosity) at varying levels of the input factor effects (e.g., fraction of coarse, medium and fine particles). During the sensitivity analysis, the complete range of each input factor is discretized into 10 subdivisions. For example, the fractions of coarse and fine ranges of 0–100% are each divided into 10 or less subdivisions, and a total of 65 predicted porosity values are obtained (Table 5). The restriction that the summation of the three powders must add up to 100% produces this number of 65 different compositions.

The utility of sensitivity analysis in this research is that it enables the researcher or engineer to easily identify the experimental conditions for lowest porosity so that problem compositions can be avoided. Results of prediction runs using the ANN model show the effects of three factors at a time on the surface plot of the porosity (Figs. 5 and 6). This ternary plot demonstrates the power of the ANN model for predictions of best packing mixture compositions. It is substantially in agreement with the 3D plot obtained by the DOE approach (Fig. 7). ■

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Table 4 Complete Data Set Used in ANN Model Construction

No.	Coarse (%) F46 D_{50} = 350	Medium (%) F320 D_{50} =30	Fine (%) F1200 D_{50} = 3	Measured porosity (%)	Predicted porosity (%)	PAE† for ANN model
1	10	90	0	42.7	43.4	1.6
2	10	80	10	40.4	39.6	1.9
3	50	40	10	27.2	28.6	5.1
4	30	50	20	31.9	31.0	2.9
5	100	0	0	34.2	35.2	2.9
6	10	0	90	44.3	44.4	0.3
7	30	10	60	36.0	35.8	0.5
8	60	30	10	27.4	27.9	1.7
9	0	0	100	45.9	45.1	1.7
10	30	30	40	30.5	30.6	0.5
11	0	90	10	41.2	41.5	0.7
12	10	60	30	34.0	34.5	1.4
13	0	30	70	31.6	35.8	13.4
14	50	0	50	32.8	32.6	0.5
15	40	10	50	32.6	32.0	2.0
16	40	30	30	27.8	28.4	2.0
17	30	20	50	33.4	32.4	3.1
18	0	100	0	44.8	44.8	0.1
19	0	50	50	35.8	34.2	4.4
20	0	0	100	45.9	45.1	1.7
21	20	20	60	35.9	35.4	1.5
22	0	50	50	36	34.2	4.9
23	10	20	70	39.6	37.3	5.8
24	70	0	30	28.9	29.4	1.8
25	20	10	70	38.4	39.0	1.4
26	30	0	70	38.6	40.6	5.2
27	40	40	20	28.8	28.7	0.5
28	90	10	0	32.5	32.5	0.1
29	10	70	20	36.3	36.4	0.2
30	40	50	10	31.6	30.4	3.7
31	50	20	30	26.3	27.5	4.4
32	30	70	0	37.4	37.6	0.5
33	50	50	0	32.9	31.9	3.1
34	90	0	10	33.8	31.9	5.6
35	20	30	50	35.1	33.2	5.4
36	10	40	50	32.9	33.9	3.0
37	20	40	40	32.1	32.3	0.7
38	70	10	20	27.3	27.8	2.0
39	20	50	30	29.8	32.5	9.0
40	66.67	16.67	16.67	27.2	27.4	0.8
41	50	10	40	29.4	29.0	1.3

Percent average absolute error (PAAE) for training of ANN model is 2.7%.

†PAE is percent absolute error.

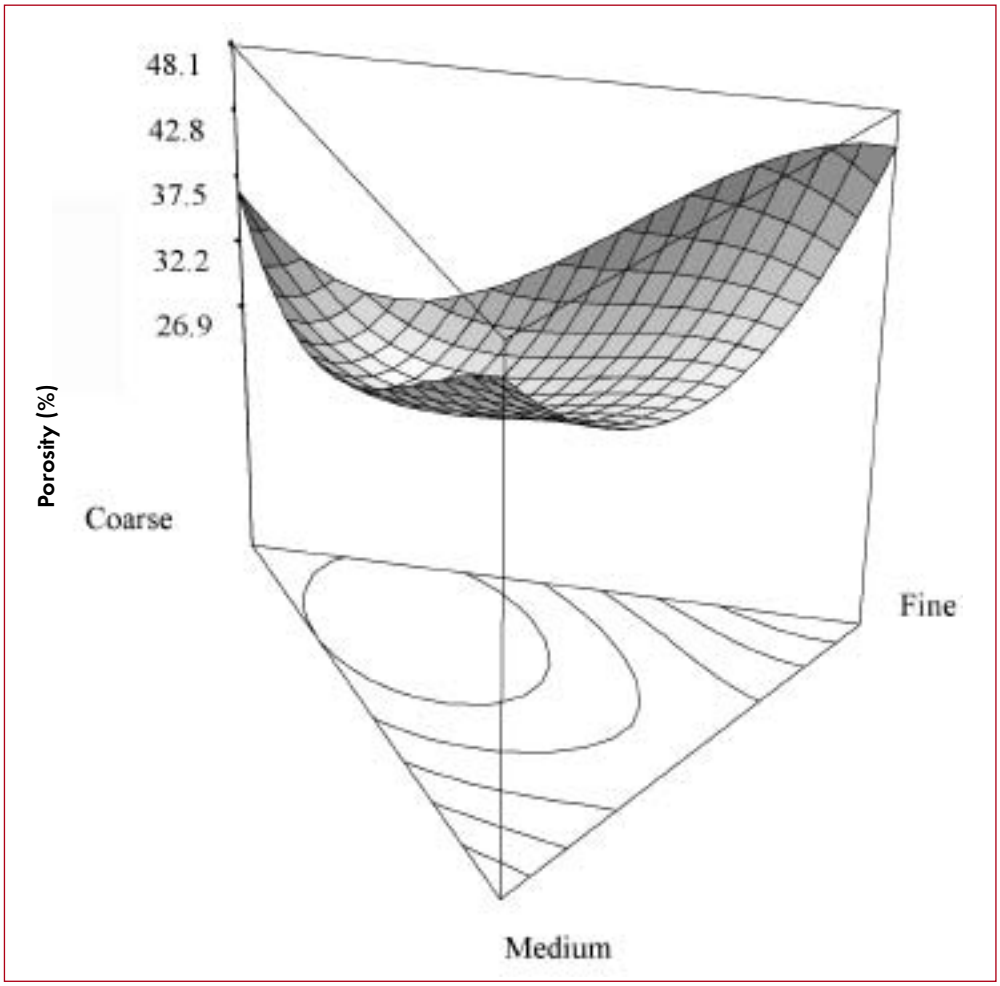

Fig. 5 Surface plot of ANN model predictions obtained after sensitivity analysis.

Table 5 Random Selection of ANN Model Sensitivity Analysis

Coarse (%) F46 D50 = 350	Medium (%) F320 D50 = 30	Fine (%) F1200 D50 = 3	Predicted porosity (%)
0	0	100	45.1
10	0	90	44.4
20	0	80	43.1
30	0	70	40.6
40	0	60	36.7
50	0	50	32.6
60	0	40	30.2
70	0	30	29.4
80	0	20	30.0
90	0	10	31.9
100	0	0	35.2
0	10	90	41.8
10	10	80	40.8
20	10	70	39.0
30	10	60	35.8
40	10	50	32.0
50	10	40	29.0
60	10	30	27.7
70	10	20	27.8
80	10	10	29.3
90	10	0	32.5
0	20	80	38.3
10	20	70	37.3
20	20	60	35.4
30	20	50	32.4
40	20	40	29.3
50	20	30	27.5
60	20	20	27.1
70	20	10	28.1
80	20	0	31.0
0	30	70	35.8
10	30	60	35.0
20	30	50	33.2
30	30	40	30.6
40	30	30	28.4
50	30	20	27.3
60	30	10	27.9
70	30	0	30.4

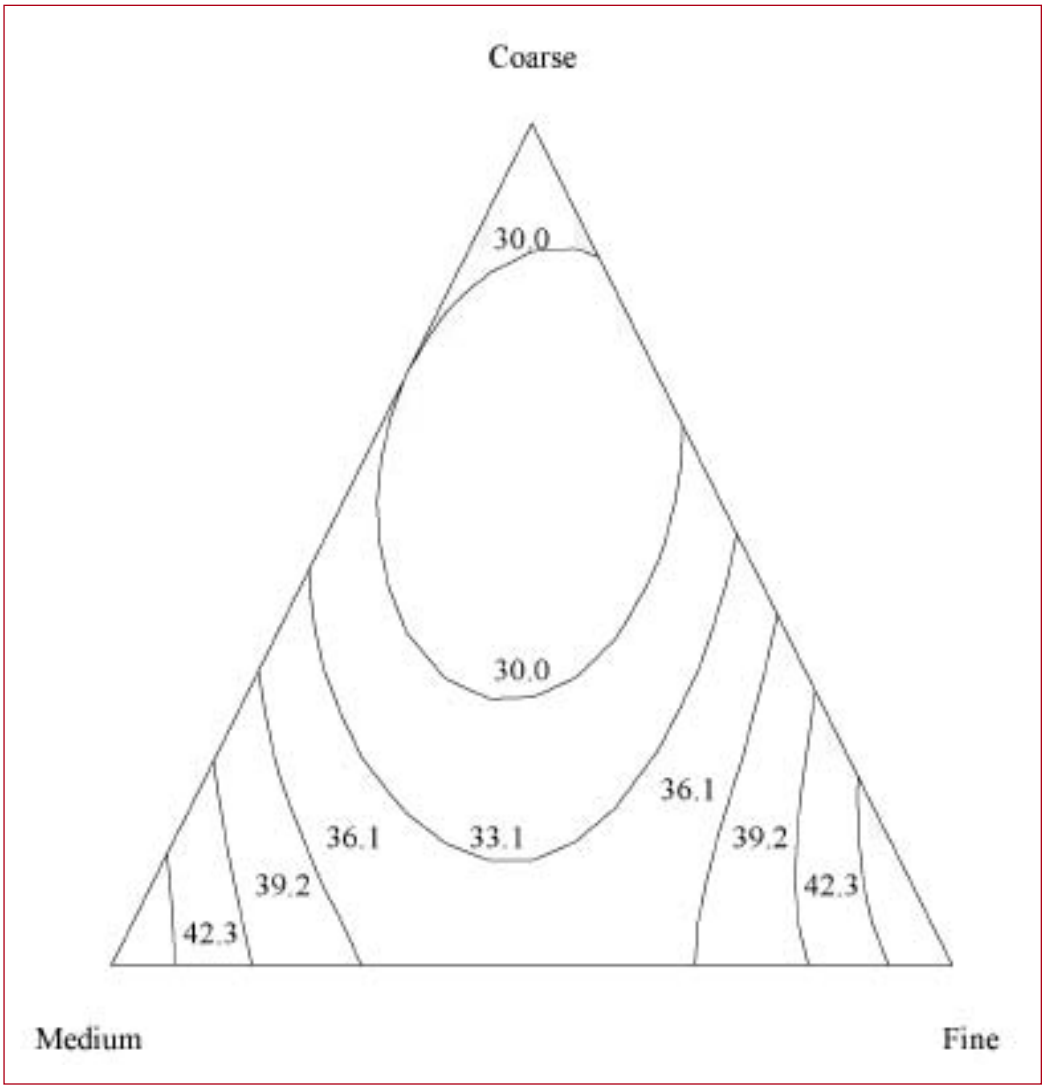

Fig. 6 ANN model predictions obtained after the sensitivity analysis. Line contours represent equal porosities.

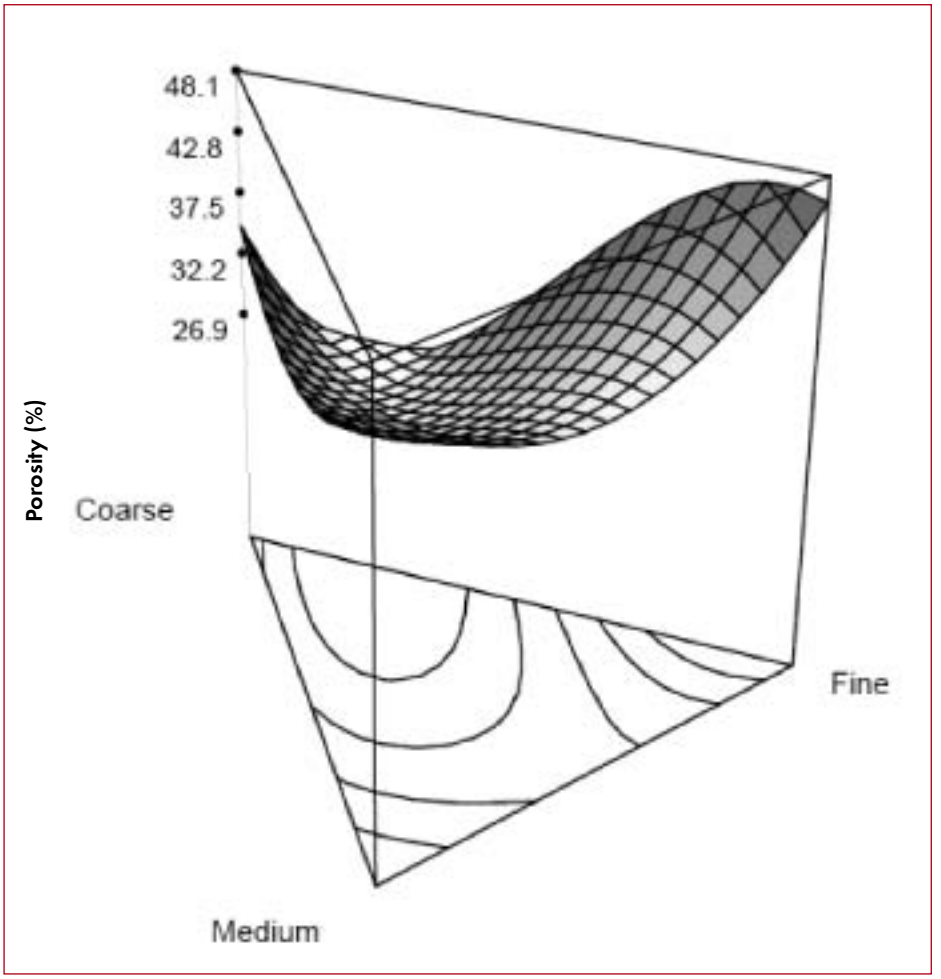

Fig. 7 Surface plot obtained using DOE model.