STATISTICAL ANALYSIS OF COMPOSITION AND TEMPERATURE FOR ALUMINA CRUCIBLE FABRICATION

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Abstract

Alumina crucibles are widely used as containers in which substances are heatedin a furnace. Slip casting is a good processing technique for producing alumina crucibles. Theproperties of alumina crucibles are dependent on the composition of their alumina slips and sintering temperatures. This research uses statistical software for an experimental design and analysis of the resulting data. Results show that the optimum composition of alumina slips for producing alumina crucibles is 900 g of alumina powder, 600 cc of deionized water, and 27 cc of Darvan C and that sintering of the work pieces should be at 1550°C.

Keywords: Alumina, crucible, statistical analysis

Introduction

Alumina or aluminum oxide (Al_2O_3) is an important raw material in ceramic industries owing to its hardness and high melting point. Aluminum oxide can naturally occur in many forms such as corundum (Al_2O_3), diaspore ($Al_2O_3 \cdot H_2O$), gibbsite ($Al_2O_3 \cdot 3H_2O$), and bauxite ($Al_2O_3 \cdot 2H_2O$). Rubies and sapphires are composed of corundum with their colors derived from trace amounts of impurities. The Bayer process, developed in 1887, is an industrial process to extract Al_2O_3 from natural ores. In this process, natural ores are washed and digested by caustic soda to yield an $NaAl(OH)_4$ solution. Then this solution is cooled resulting in precipitation of aluminum hydroxide ($Al(OH)_3$). Finally, aluminum oxide is produced when aluminum hydroxide is fired to 980°C.

Alumina exists in 3 forms: alpha, gamma, and beta. Almost all alumina occurring in the ambient atmosphere is alpha alumina. Its structure is hexagonal and close-packed. Pure alumina has a melting point of 2054°C, a density equal to 3.97 g/cm^3, a flexural strength of 410 MPa, a no-load shape stability at 1750°C, and a hardness equal to 9 on the Mohs scale (Powpan, 2012). These excellent properties result in alumina being widely used for many industrial including the manufacture of abrasives and refractory materials. One of the applications in the refractory industry is the manufacture of crucibles. Alumina

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crucibles can be very easily fabricated by a slip casting process. The properties of alumina crucibles depend largely on their slip compositions and sintering temperatures. Generally, alumina slip composition consists of 3 major components. These are alumina powder, a deflocculant, and deionized water.

There are many types of alumina powder available onthe market. The applications alumina powder used in this work is alumina A-5M (Suzhou Dexin Advanced Ceramics Co., Ltd., Suzhou, China). It is 95% pure and its average particle size is 5 μm. When alumina powder is added to deionized water, the surface of the alumina power has a positive charge because alumina has an isoelectric point at pH 8-9. Therefore an anionic electrolyte is used in the alumina slip as a deflocculant in order to inhibit the alumina powder from setting in the slip (Srilomsak, 2006a, 2006b). A common commercial anionic electrolyte is Darvan® (R.T. Vanderbilt Co., Inc., Norwalk, CT, USA) (Vanderbilt, 2012). There are several types of Darvan® for different applications. In the current work Darvan C was selected. The sintering temperature also plays a significant role in the crucible properties; hence this research examined the sintering temperatures as well.

The experiment was planned and conducted using statistical experimental design software, Design-Expert® Version 8 (Stat-Ease, Inc., Minneapolis, MN, USA). This work examined the effects of 2 independent factors (A and B). Thus a general factorial design was used in the software experimental design. The dependent factors in this study were the alumina crucible properties, namely apparent porosity, water absorption, bulk density, andlinear firing shrinkage. The purpose of this study was to determine the optimum level of the Darvan C addition and the optimum sintering temperature to produce alumina crucibles with the lowest apparent porosityandwater absorption, and the highest bulk density.

Materials and Method

The general factorial design in Design-Expert® was used in this experimental design. There were 2 independent factors, the Darvan C volume (Factor A) and sintering temperature (Factor B). There were 3 levels of Darvan C (12, 21, and 30 cc) and 3 temperature levels (1350, 1450, and 1550°C) for each replicate treatment combination. Thus, there were 3×3 or 9 combinations. Since experimental error may have been present, it was desirable to make 3 observations for each treatment combination. Therefore, there were 3×9 or 27 observations for the entire experiment. These conditions are shown in columns 1-4 of Table 1. The first column in this Table under the heading, Std, represents a standard number which is ordered according to the Darvan C volume and sintering temperature. The second column under the heading, Run, is a run number which is the order inwhich data was collected. It is important to note that the run number was selected randomly. This means that the experiments defined by the standard number were performed in random order. This randomization is essential to minimize the effects of extraneous factors that may be present. The third and fourth columns of Table 1 are the Darvan C volume and sintering temperature in each experimental observation.

Alumina (900 g) and deionized water (600 cc) were measured. Alumina powder, deionized water and Darvan C were mixed according to the run numbers in Table 1. The mixture was ball milled for 2 days to obtain material for alumina slips. The alumina slips were cast in plaster molds and allowed to set for 15 min. Then the slips were drained out of their molds. After 1 day, the work pieces were taken out of their plaster molds. The work pieces were dried at 60°C in an oven for 2 days. The diameters of the dried alumina crucibles were measured. The dried work pieces were sintered at the temperatures specified in the fourth column of Table 1. The following heat and cooling profile was used:

a. Samples were heated from room temperature to 550°C at a rate of 1°C/min.

b. The sample temperature was held at 550°C for 3 h.

c. Samples were heated from 550°C to the temperature specified in the fourth column of Table 1. A heating rate of 2°C/min was used. Upon reaching the specified temperature, this temperature was maintained for 2 h.

d. Cooling to room temperature was done at a rate 5°C/min.

The work pieces were removed from the furnace. Their diameters were determined and

Table 1. Standard number (Std), Run number (Run), Darvan volume (Darvan), Sintering temperature (Temp), Apparent porosity (Porosity), Water absorption (Absorption), Bulk density (Density) and Linear firing shrinkage (Shrinkage)

Std	Run	Factor 1 A:Darvan (cc)	Factor 2 B:Temp (°C)	Response 1 porosity (%)	Response 2 absorption (%)	Response 3 density (g/cm^3)	Response 4 shrinkage (%)
1	26	12	1350	34.51	13.97	2.47	2.81
2	23	12	1350	34.28	13.80	2.48	2.35
3	14	12	1350	33.85	13.58	2.49	2.48
4	19	21	1350	34.44	13.65	2.52	2.76
5	7	21	1350	34.24	13.52	2.53	3.36
6	16	21	1350	33.76	13.23	2.55	3.08
7	21	30	1350	33.67	13.20	2.55	3.19
8	9	30	1350	33.74	13.16	2.56	2.29
9	6	30	1350	33.32	12.92	2.58	3.65
10	20	12	1450	31.05	11.92	2.61	4.76
11	5	12	1450	32.13	12.48	2.58	3.57
12	12	12	1450	31.65	12.25	2.58	5.36
13	1	21	1450	30.25	11.26	2.69	5.25
14	8	21	1450	29.40	10.82	2.72	5.85
15	2	21	1450	30.39	11.35	2.68	4.64
16	4	30	1450	30.17	11.17	2.70	3.97
17	25	30	1450	29.98	11.07	2.71	6.34
18	27	30	1450	30.01	11.08	2.71	4.53
19	17	12	1550	23.96	8.20	2.92	8.58
20	18	12	1550	25.59	9.11	2.81	7.36
21	13	12	1550	25.26	9.00	2.81	7.02
22	11	21	1550	24.56	8.47	2.90	8.14
23	15	21	1550	24.24	8.32	2.91	7.52
24	24	21	1550	24.47	8.44	2.90	6.34
25	10	30	1550	25.18	8.70	2.89	8.52
26	22	30	1550	24.54	8.50	2.89	8.70
27	3	30	1550	24.82	8.62	2.88	10.05

linear firing shrinkages calculated usingthe following equation:

$$Linear\ firing\ shrinkage\ (\%) = \frac{(Dried\ length - Fired\ length)}{Dried\ length} \times 100$$

Apparent porosity, water absorption, and bulk density were determined according to ASTM C373-88 (ASTM, 1994). Dependent factors (apparent porosity, water absorption, bulk density, and linear firing shrinkage) were examined using Analysis of Variance (ANOVA) in Design-Expert®. This was done to determine if the independent factors (the Darvan C volume and sintering temperature) and their interactions had significant effects (>95% confidence) on the dependent factors. Regression equations were developed to predict the effects of the Darvan C additions and sintering temperature on apparent porosity, water absorption, and bulk density of the alumina crucibles. ANOVA assumptions were verified with normality and residual versus predicted values plots.

Results and Discussion

Figure 1 shows the alumina crucibles fabricated in this research. The fifth through eighth columns of Table 1 show the alumina crucible properties (i.e., apparent porosity, water absorption, bulk density, and linear firing shrinkage) as a function of the Darvan C volume and sintering temperature. Analysis of the effect of the independent factors (the Darvan C volume and sintering temperature) and their effects on the dependent factors (apparent porosity, water absorption, bulk density, and linear firing shrinkage) by ANOVA yields the following results.

a. Apparent Porosity

Table 2 shows the ANOVA results for apparent porosity. The Darvan C volume (A), sintering temperature (B), and square of the sintering temperature (B^2) have significant effects (more than 95% confidence) on the apparent porosity of the alumina crucibles. The equation for predicting the apparent porosity is:

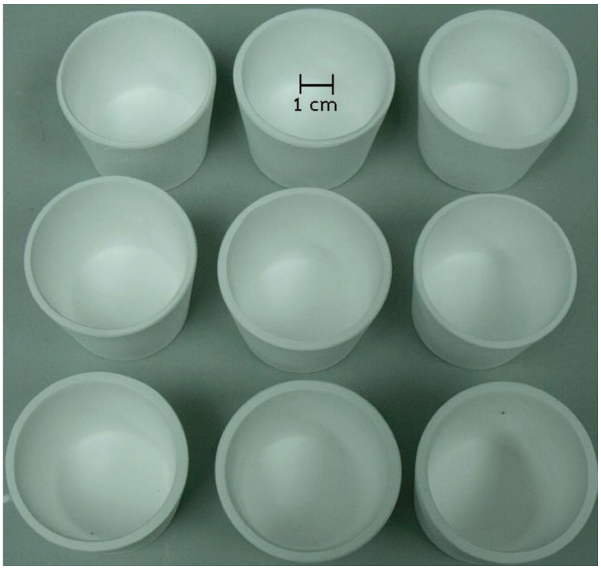

Figure 1. Alumina crucibles fabricated in this experiment,the sizes of which are estimated at 4.5 cm diameter and 5 cm height

Apparent porosity (%)

$$= -154.31 - 0.04 \times \text{Darvan (cc)} + 0.30 \times \text{Temp } (^{o}\text{C}) - 1.20 \times 10^{-4} \times \text{Temp}^{2} (^{o}\text{C}^{2})$$

Figure 2 is a normal plot of the residual values to check the ANOVA normality assumption. The plot slightly diverges from a straight line. However, this divergence is small. Therefore the assumption that the residuals are normally distributed and have a mean ~0 is satified (Montgomery, 2001). Figure 3 shows the residual versus predicted apparent porosity plot. This plot is used for checking the ANOVA assumption for the homoginety of variance. When the variance is homegeneous, the residual values are dispersed evenly around the zero line (Montgomery, 2001). Figure 3 has this characteristic implying that the homogeneity of variance assumption is met. Three dimensional and contour plots of the predicted apparent porosity as a function of the Darvan C volume and sintering temperature are shown in Figures 4 and 5. From both graphs it is clear that the lowest apparent porosity was obtained by using the highest Darvan C volume in the alumina slip and sintering the crucible at 1550°C.

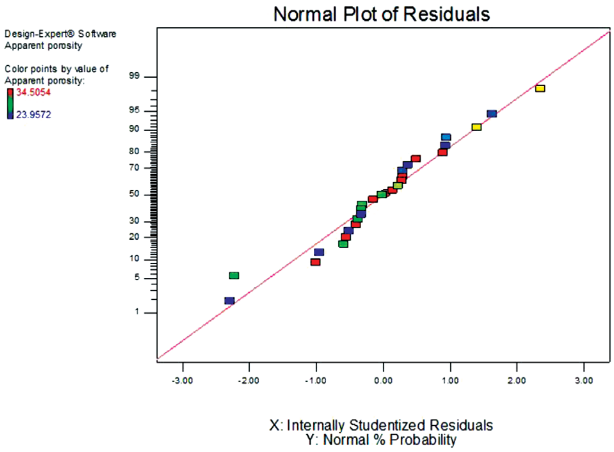

Figure 2. Normal probability plotof residuals for apparent porosity of alumina crucibles

Table 2. Analysis of Variance (ANOVA) for apparent porosity data

Source	Sum of squares	df	Mean square	F value	p-value Prob > F	
Model	395.68	3	131.89	429.17	< 0.0001	significant
A-Darvan C	2.61	1	2.61	8.49	0.0078	
B-Temperature	384.39	1	384.39	1250.79	< 0.0001	
B^2	8.67	1	8.67	28.22	< 0.0001	
Residual	7.07	23	0.31			
Cor Total	402.75	26				

Note: Definitions of Sum of Squares, df, F Value, p-value Prob> F are in Montgomery (2001) and Anderson and Whitcomb (2007)

b. Water Absorption

Table 3 displays the ANOVA results for water absorption. The Darvan C volume (A), sintering temperature (B), and square of the sintering temperature (B^2) have significant effects on the water absorption of the alumina crucibles. A regression equation to predict the water absorption is:

$$\text{Water absorption (\%)} = -48.92 - 0.17 \times \text{Darvan (cc)} + 0.11 \times \text{Temp (}^o\text{C)} + 3.15 \times 10^{-3} \times \text{Darvan}^2 \text{ (cc}^2\text{)} - 4.64 \times 10^{-5} \text{ Temp}^2 \text{ (}^o\text{C}^2\text{)}$$

The normal probability and studentized residual versus predicted water absorption values were plotted but are not shownhere. Both plots indicate normality. The ANOVA assumptions were satisfied. Figures 6 and 7 are 3D surface and contour plots of the predicted water absorption versus the Darvan C volume and sintering temperature. From both Figures it can be seen that the optimun conditions to produce the alumina crucibles with the lowest water absorption is by using 27 cc Darvan C and sintering at 1550^{o}C.

c. Bulk Density

Table 4 shows the ANOVA results for bulk density. The Darvan C volume (A), sintering temperature (B), square of the Darvan C volume (A^2), and square of the sintering temperature (B^2) have significant

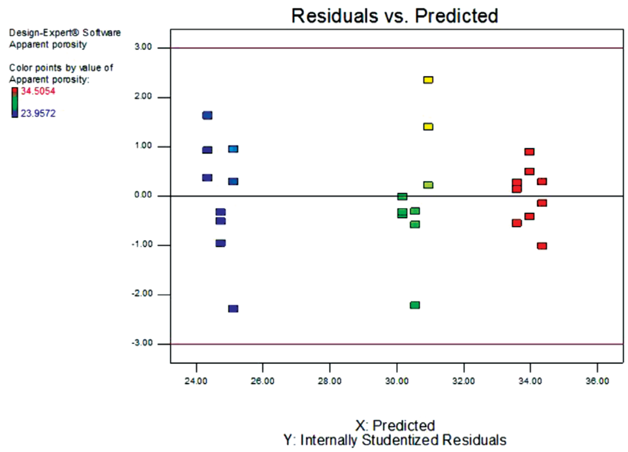

Figure 3. Plot of studentized residuals versus predicted apparent porosity

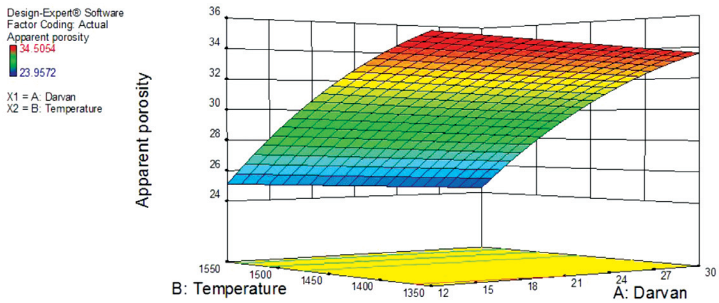

Figure 4. Three dimensional (3D) surface plot of predicted apparent porosity as a function of the Darvan C volume and sintering temperature

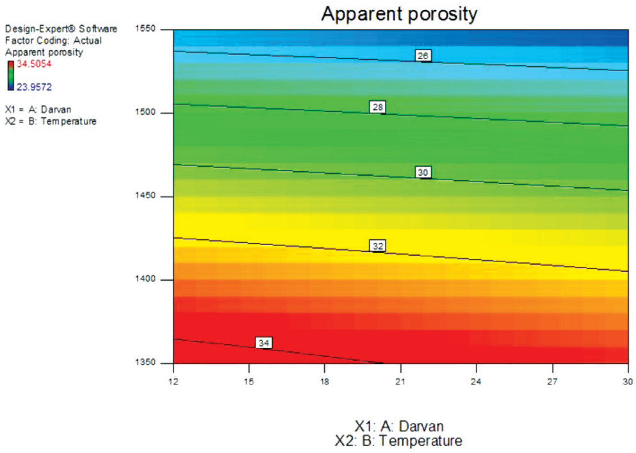

Figure 5. Contour plot of predicted apparent porosity as a function of the Darvan C volume and sintering temperature

Table 3. Analysis of Variance (ANOVA) for water absorption data

Source	Sum of squares	df	Mean square	F value	p-value Prob > F	
Model	109.59	4	27.40	296.73	< 0.0001	significant
A-Darvan C	1.92	1	1.92	20.83	0.0002	
B-Temperature	105.98	1	105.98	1147.86	< 0.0001	
A^2	0.39	1	0.39	4.24	0.0516	
B^2	1.29	1	1.29	13.99	0.0011	
Residual	2.03	22	0.09			
Cor Total	111.62	26				

Table 4. Analysis of Variance (ANOVA) for bulk density data

Source	Sum of squares	df	Mean square	F value	p-value Prob > F	
Model	0.56	4	0.14	478.92	< 0.0001	significant
A-Darvan C	0.04	1	0.04	125.40	< 0.0001	
B-Temperature	0.48	1	0.48	1666.67	< 0.0001	
A^2	0.01	1	0.01	29.94	< 0.0001	
B^2	0.01	1	0.01	22.97	< 0.0001	
Residual	0.01	21	0.00			
Cor Total	0.56	25				

Note: This analysis does not include the data on standard number 19 (run number 17) because it is, outlier data, in other words this data is much too different from other data to be used in statistical analysis effects on the bulk density of the alumina crucibles. A regression equation for predicting the bulk density is:

Bulk density (g/cm^3)
= 7.00 + 0.03 × Darvan (cc)- 0.01 × Temp (°C) - 4.75 × 10 - 4 × Darvan^2(cc^2) + 3.37 × 10^{-6} × Temp^2 (°C^2)

There is no abnormality in the normal probability and studentized residual versus predicted bulk density plots. The ANOVA assumptions were satisfied. Consequently, the ANOVA results are acceptable. Figures 8 and 9 are 3D surface and contour plots of the predicted bulk density versus the Darvan C volume and sintering temperature. From both Figures it can concluded that the optimumconditions to produce the highest density alumina crucibles occurred when using 27 cc Darvan C and sintering at the highest temperature.

d. Linear Firing Shrinkage

The normal probability plot of the residual of linear firing shrinkage has an "S" shape and the studentized residual versus predicted linear firing shrinkage has a funnel shape.Therefore the ANOVA assumptions

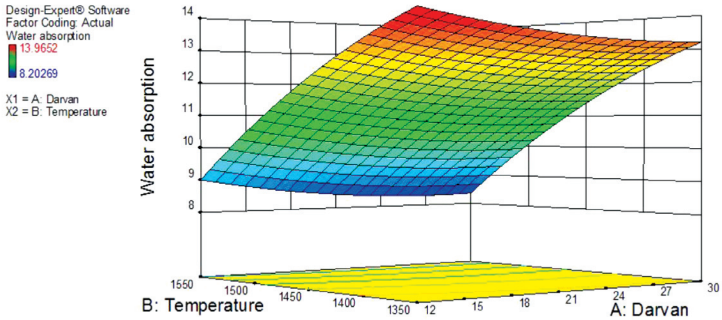

Figure 6. Three dimensional (3D) surface plot of predicted water absorption as a function of the Darvan C volume and sintering temperature

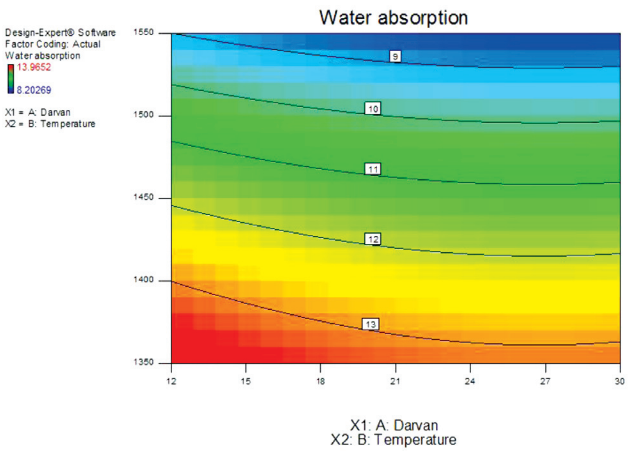

Figure 7. Contour plot of predicted water absorption as a function of the Darvan C volume and sintering temperature

are not satisfied. Data were transformed as recommended by the software and the ANOVA analysis was repeated. Table 5 shows the ANOVA result for the linear firing shrinkage data after the transformation. Only the sintering temperature (B) has a significant effect on the linear firing shrinkage of the alumina crucibles. The resulting regression equation for predicting thelinear firing shrinkageis:

$$\text{Log10 (\% linear firing shrinkage)} = -2.62 + 3.31 \times 10^{-3} \times \text{Darvan (cc)} + 2.23 \times 10^{-3} \times \text{Temp (}^{o}\text{C)}$$

The normal probability and residual versus predicted linear firing shrinkage of the transformed data were plotted. No abnormality was observed; hence, the ANOVA results for the transformed data are acceptable. Figures 10 and 11 are 3D surface and contour plots of the predicted linear firing shrinkage versus the Darvan C volume and sintering temperature. From both Figures it can be seen that the linear firing shrinkage is decreased by reducing the Darvan C volume and sintering temperature.

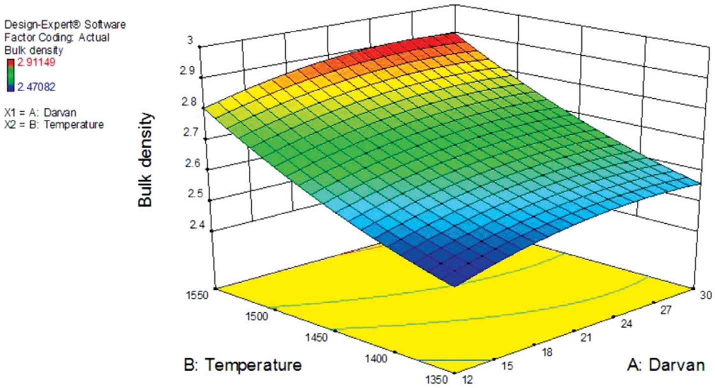

Figure 8. Three dimensional (3D) surface plot of predicted bulk density as a function of the Darvan C volume and sintering temperature

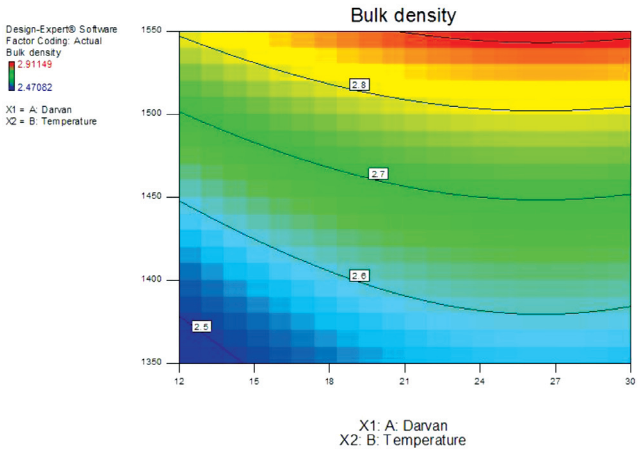

Figure 9. Contour plot of predicted bulk density as a function of the Darvan C volume and sintering temperature

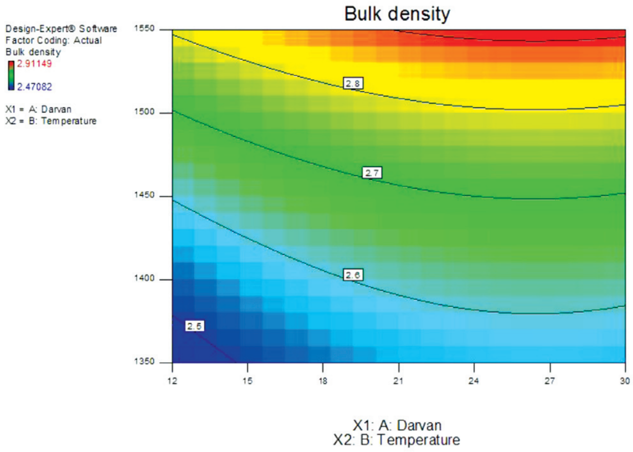

Figure 10. Three dimensional (3D) surface plot of predicted linear firing shrinkage as a function of the Darvan C volume and sintering temperature

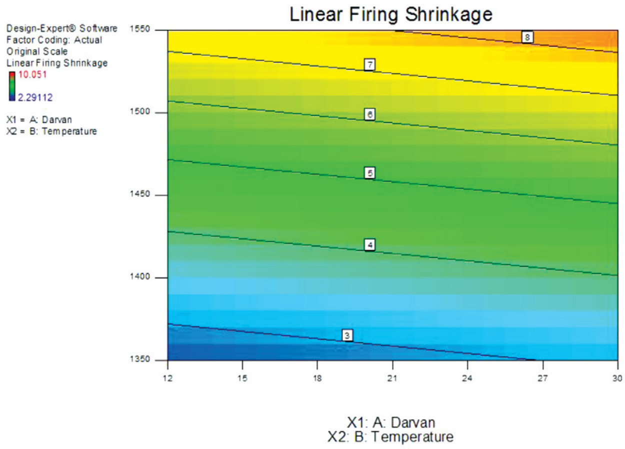

Figure 11. Contour plot of predicted linear firing shrinkage as a function of the Darvan C volume and sintering temperature

Table 5. Analysis of Variance (ANOVA) for transformed linear firing shrinkage

Source	Sum of squares	df	Mean square	F value	p-value Prob> F	
Model	0.91	2	0.46	106.56	< 0.0001	significant
A-Darvan C	0.02	1	0.02	3.73	0.0652	
B-Temperature	0.89	1	0.89	209.38	< 0.0001	
Residual	0.10	24	0.00			
Cor Total	1.01	26				

Note: Transformation was done by using equation linear firing shrinkage = log (linear firing shrinkage)*

Conclusions

This work used Design-Expert® software to design and analyze the effects of the Darvan C volume and sintering temperature upon apparent porosity, water absorption, and bulk density of alumina crucibles. It is evident that the optimum composition of the alumina slips for producing the lowest porosity (23.96%) and water absorption (8.20%), and the highest density (2.92 g/cm^3) alumina crucibles is 900g of alumina powder, 600 cc of deionized water, and 27 cc of Darvan C and that the sintering of the work pieces should be at 1550^{o}C.

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