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COMPOSITION OPTIMIZATION OF ALUMINA SUSPENSIONS WHICH CONTAIN WASTE ALUMINA POWDER

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Abstract

This paper reports the results of studying the stabilization of alumina ceramics suspensions which contain a considerate amount of waste alumina powder. The study focused on the investigation of the influence of different amounts of the commercial dispersant Tiron, binder PVA - poly(vinyl alcohol) and the magnesium spinel as additives for the stabilization of highly concentrated alumina suspensions with 20 wt. % of waste (or secondary) alumina powder. Waste alumina powder is obtained after the green machining in factory production of ceramics. Suspensions with different amount of dispersant, binder and additive were prepared based on the response surface methodology design to optimize the suspension viscosity. The viscosity was used for the suspension stability estimation because it is very simple, rapid and accurate method. The lowest viscosity value of all suspensions was achieved with the addition of 0.05 % Tiron, 0.1 % PVA and 0.2 % magnesium spinel expressed on dry weight basis. The obtained results suggest the waste alumina powder is appropriate for shaping new ceramic products by slip casting method.

Keywords: alumina suspensions; waste alumina; response surface methodology; slip casting

1. Introduction

The depletion of natural resources impose a serious problem to the modern society. The awareness of the need for sustainable development in terms of safe reuse of industrial waste is becoming increasingly popular among researchers. The transformation of this waste into valuable materials is emerging as a possible solution to reduce the environment pollution. The reuse of recovered waste generated from the manufacturing processes involves the need for recycling as secondary raw materials [1], [2], [3]. An aspect that is particularly important when dealing with a problem related to waste is their environmental impact, although the economic benefits gained from waste recycling must not be ignored [4].

2. Literature Review

Ceramic materials are already a well-known and widely used group of technical materials. These materials have convenient properties such as high density and strength, high hardness as well as high temperature and corrosion resistance. These properties provide the ability to use the ceramic materials for many technical purposes [5], [6].

During the green machining of the green body, a certain amount of waste ceramic powder is generated which typically remains unused. In addition, the waste ceramic powder should be disposed as non-hazardous waste in a legally prescribed manner. Some alternatives have been searched to refine alumina ceramics microstructure by changing their composition. Li et al. [7] investigated the sintering of alumina powder compacts doped with various amounts of titanium dioxide and cupric oxide mixtures prepared at a weight ratio of 4:1. Compacts doped with 5 wt. % of such mixture showed sufficient mechanical properties after low-temperature sintering. Marie et al. [8] studied bio-based alumina concentrated suspensions with low environmental impact were prepared from bio-polymer additives. The study of the rheological and chemical characteristics of the suspensions has been showed that the incorporation of the additives has a strong role on the final characteristics of the alumina suspensions.

Slip casting as colloidal shaping method has been suggested as method to produce high quality ceramic green body. The first step in these methods is the preparation of a well-dispersed low viscosity suspension loaded with high amounts of solids to facilitate the mold filling process. The rheological properties of the concentrated suspensions have a key role in controlling the shape forming behaviour and subsequently, green body properties. The stability of a colloidal suspension is reflected in the properties of the green compact, which also affects the properties of the sintered material [9], [10], [11]. The preparation of alumina ceramic suspensions with the addition of waste ceramic powder has not been reported so far, to the author's best knowledge. The purpose of this study is to prepare high-load stable alumina suspensions which contain 20 % of waste alumina expressed on dry weight basis (dwb %) of alumina powders. Response surface methodology (RSM) was used to optimize different weight additions of the dispersant Tiron, binder PVA and the magnesium spinel on the viscosity of prepared alumina suspensions. Response surface models were established based on experimental data. Response surface methodology (RSM) was first proposed in 1951 and includes conducting of experiments, modeling and data analysis [12],[13].

3. Materials and Methods

Waste alumina powder which is obtained after green machining and high-purity Al_2O_3 powder with the average particle size 300-400 nm (Alcan Chemicals, USA) were used in this study. Mixtures of $Al_2O_3 - Al_2O_3$ (waste) with composition of 20 wt. % waste alumina powder were prepared. A commercial dispersant Tiron (4,5-dihydroxy-1,3-benzenedisulfonic acid disodium salt monohydrate) manufactured by Sigma-Aldrich Chemie GmbH, Germany was used to stabilize highly concentrated alumina suspensions [14] ,[15]. The binder PVA manufactured by Sigma-Aldrich Chemie GmbH, Germany [16] was added to the ceramic suspension in order to improve the strength of the green bodies. Magnesium spinel (magnesium aluminium oxide) manufactured by Alfa Aesar, USA was used to inhibit the abnormal alumina grain growth during the sintering process of alumina green bodies [17], [18].

3.1. Slip Preparation

The mixture of dry alumina powders was prepared previously by mixing 20 dwb % (19.2 g) of waste alumina powder versus 80 dwb % (76.8 g) of high-purity alumina powder. The mixture of dry alumina powders and different amounts of dispersant, binder and magnesium spinel were mixed with deionized water (40 mL) and added into the grinding jar of the planetary ball mill PM 100 (Retsch GmbH, USA) to prepare 70 wt. % alumina suspensions (Table 1). Ten alumina balls were added into the grinding jar and used for the mixture homogenization. The homogenization lasted for 90 minutes at a speed of 300 rpm. The wear resistance of the used alumina balls is high and the assumption is that the impact of possible alumina balls wear debris in stated conditions of mixing is negligible [19]. Alumina balls were separated from the suspension after the homogenization using a strainer. The suspension underwent an ultrasonic treatment for 15 min in an ultrasonic bath – BRANSONIC 220 (Branson Ultrasonics Corp., USA) to remove the air bubbles and achieve better homogeneity.

3.2. Determination of Rheological Properties

Rheological properties were determined using a rotational viscometer DV-III Ultra (Brookfield Engineering Laboratories, Inc., USA) in a small sample chamber with spindle SC4-18 for lower values of viscosity and with spindle SC4-34 for higher values of viscosity. Pre-shearing has been performed for 2 min at a shear rate of 100 s^{-1} followed by a 2 min rest period in order to provide a common and consistent shear history for the system. The shear rate of 50 s^{-1} was set because that is the shear rate usually achieved during the gravity slip casting. The suspension stability was estimated by the measurements of viscosity for all prepared suspensions. Temperature was kept constant at 25±1 °C using a thermostatic bath Lauda EcoRE 415 (LAUDA-Brinkmann, LP, USA).

4. Results and Discussions

4.1. Modeling

The value range of each factor was determined according to previous tests. The range of dispersant Tiron was from 0.5 to 0.15 dwb %, the range of binder PVA was from 0.1 to 0.5 dwb % and the range of magnesium spinel was from 0.2 to 1 dwb % expressed on dry weight basis of alumina powder. These 3 factors with 3 levels gave a total of 15 experiments in randomized order, as per Design Expert software Box-Behnken response surface design.

The corresponding response viscosity of each prepared suspension was measured (Table 1). Then, the RSM was used to find out the optimal value of each factor to obtain minimum viscosity.

Run	Factor 1 *A*: Tiron, dwb%	Factor 2 *B*: PVA, dwb %	Factor 3 *C*: Magnesium spinel, dwb %	Response viscosity, mPa · s	Predicted viscosity, mPa · s
1	0.1	0.1	1	40.72	38.06
2	0.05	0.3	0.2	51.03	47.79
3	0.05	0.3	1	52.22	52.45
4	0.15	0.3	1	55.34	58.58
5	0.1	0.5	0.2	95.51	98.16
6	0.15	0.5	0.6	107.75	105.33
7	0.1	0.3	0.6	54.68	56.33
8	0.1	0.3	0.6	57.76	56.33
9	0.15	0.1	0.6	42.79	42.20
10	0.05	0.5	0.6	91.26	91.86
11	0.1	0.5	1	100.41	99.59
12	0.1	0.3	0.6	56.53	56.33
13	0.1	0.1	0.2	38.33	39.15
14	0.05	0.1	0.6	32.02	34.44
15	0.15	0.3	0.2	63.11	62.89

Table 1. Experimental results

The predicted viscosity (Table 1) was obtained using the second order polynomial equation by applying multiple regression analysis on the experimental data, the following quadratic equation is obtained which describes the relation between the dependent and the independent variables:

$$\text{Viscosity (mPas)} = 24.91878 + 177.86637 * A - 59.1614 * B + 11.37873 * C + 142.834390 * A * B - 112.01277 * A * C + 7.86044 * B * C - 236.73331 * A^2 + 318.05723 * B^2 - 1.93004 * C^2 \quad (1)$$

Where:
- A is the weight ratio of the dispersant Tiron expressed on dry weight basis (dwb %) of alumina powder,
- B is the weight ratio of the binder PVA (dwb %),
- C is the weight ratio of magnesium spinel (dwb %).

By analysing the ANOVA data for the response surface quadratic model of suspension's viscosity (Table 2), the higher model F-value (84.05) and the associated lower p-values (p<0.0001) demonstrated that the polynomial regression models were suitable to determine the optimum composition of selected additives for the preparation of alumina suspensions, which contain 20 dwb. % waste alumina powder with reasonably low viscosity to facilitate the mold filling process. The p-value of the variable B (weight ratio of PVA) was less than 0.0001 indicating that the change of the binder content had a significant effect on the viscosity of the prepared alumina suspensions. The remaining two variables (content of Tiron and magnesium spinel) had a relatively fewer effect on the obtained viscosity because their p-values were above 0.0001. High R^2 (0.9934), adjusted R^2 (0.9816) and predicted R^2 (0.9030) values indicate that the variation could be accounted for by the data satisfactorily fitting the model. The coefficient of variation (*C.V.* %) of less than 10 % clearly exhibits very high degree of precision and good reliability of the conducted experiments.

After setting up the minimum viscosity as for the optimization, with independent variables set in previously defined range, the optimum solution showed a desirability of 0.994. The optimum values were as follows: 0.05 % Tiron, 0.1 % PVA and 0.2 % magnesium spinel expressed on dry weight basis. Fig. 1 represents the response surface plots (3D) showing the effects of variables A: Tiron (dwb %); B: PVA (dwb %); C: Magnesium Spinel (dwb %) on the response viscosity of the prepared suspensions.

Source	Sum of Squares	df	Mean Square	*F*-Value	*p*-value
Model	8132.05	9	903.56	84.05	< 0.0001
A (Tiron)	225.42	1	225.42	20.97	0.0060
B (PVA)	7264.81	1	7264.81	675.76	< 0.0001
C (Mg Spinel)	0.062	1	0.062	5.738E-003	0.9426
AB	8.16	1	8.16	0.76	0.4234

AC	20.07	1	20.07	1.87	0.2300
BC	1.58	1	1.58	0.15	0.7171
A^2	1.29	1	1.29	0.12	0.7428
B^2	597.62	1	597.62	55.59	0.0007
C^2	0.35	1	0.35	0.033	0.8635
Residual	53.75	5	10.75		
Pure Error	4.80	2	2.40		
Cor. Total	8185.80	14		R^2	0.9934
Std. Dev.	3.28			Adjusted R^2	0.9816
Mean	62.63			Predicted R^2	0.9030
C.V. %	5.24			Adequate Precision	26.478

Table 2. Analysis of variance for response surface quadratic model of suspension's viscosity

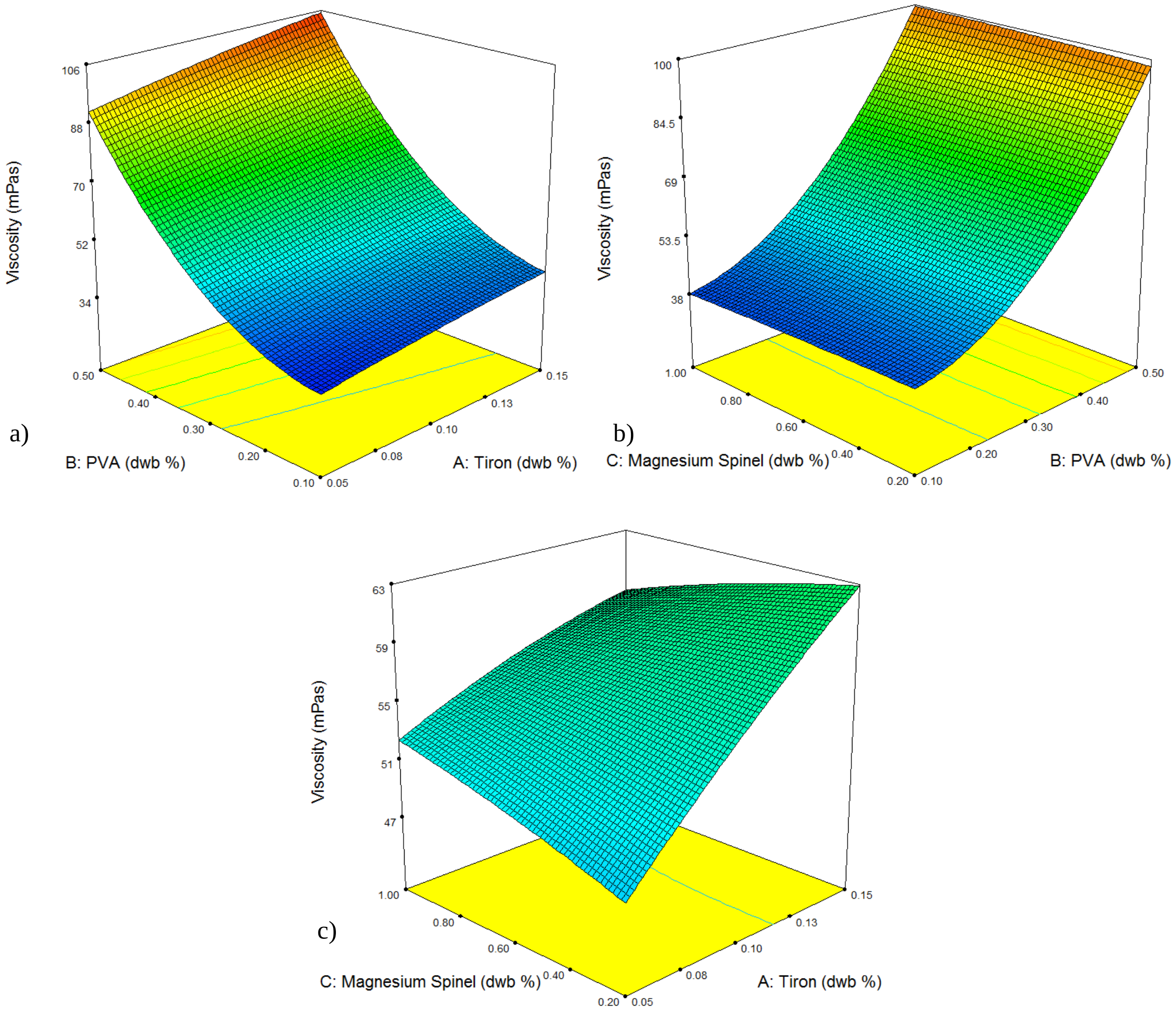

Fig. 1. a) 3D response surface for the effect of PVA and Tiron on suspensions viscosity at constant content of magnesium spinel (0.6 dwb %), b) 3D response surface for the effect of PVA and magnesium spinel on suspensions viscosity at constant content of Tiron (0.1 dwb %), c) 3D response surface for the effect of magnesium spinel and Tiron on suspensions viscosity at constant content of PVA (0.3 dwb %)

4.2. Effect of Dispersant Tiron

Studies on dispersing alumina particles using the effect of the molecular structure of low molecular weight organic dispersants such as Tiron for the preparation of highly concentrated stable alumina suspensions have been reported [20], [21].

It is crucial to determine the optimum amount of the dispersant required for the minimum viscosity to obtain stable alumina suspensions. Any percentage higher or lower than the optimum range will increase the viscosity which will in turn result with the instability of suspensions. An increase in Tiron content for the investigated range has shown a slightly impact on increase of viscosity. The *p*-value of this variable was 0.0060 (Table 1 and Fig. 2a).

4.3. Effect of Binder PVA

Poly(vinyl alcohol) is a hydrosoluble organic polymeric binder which is added to ceramic suspension in order to improve the strength of the green bodies [16]. It was observed that the viscosity increases with increase of PVA weight ratio. Despite the fact that increasing weight ratio PVA has the positive effect on the strength of the green bodies, increasing content of PVA negatively affect the stability of prepared suspensions by increasing viscosity. The *p*-value of the variable was less than 0.0001 indicating that the change of the binder content had a significant effect on the viscosity of the prepared alumina suspensions (Table 1 and Fig. 2b).

4.4. Effect of Magnesium Spinel

Magnesium spinel is usually added to inhibit abnormal alumina grain growth during sintering process of alumina green bodies [18]. Upon increasing the content of magnesium spinel is shown no effect on viscosity in the investigated range. This statement is also confirmed by the *p*-value which was above 0.0001 (Table 2 and Fig. 2c).

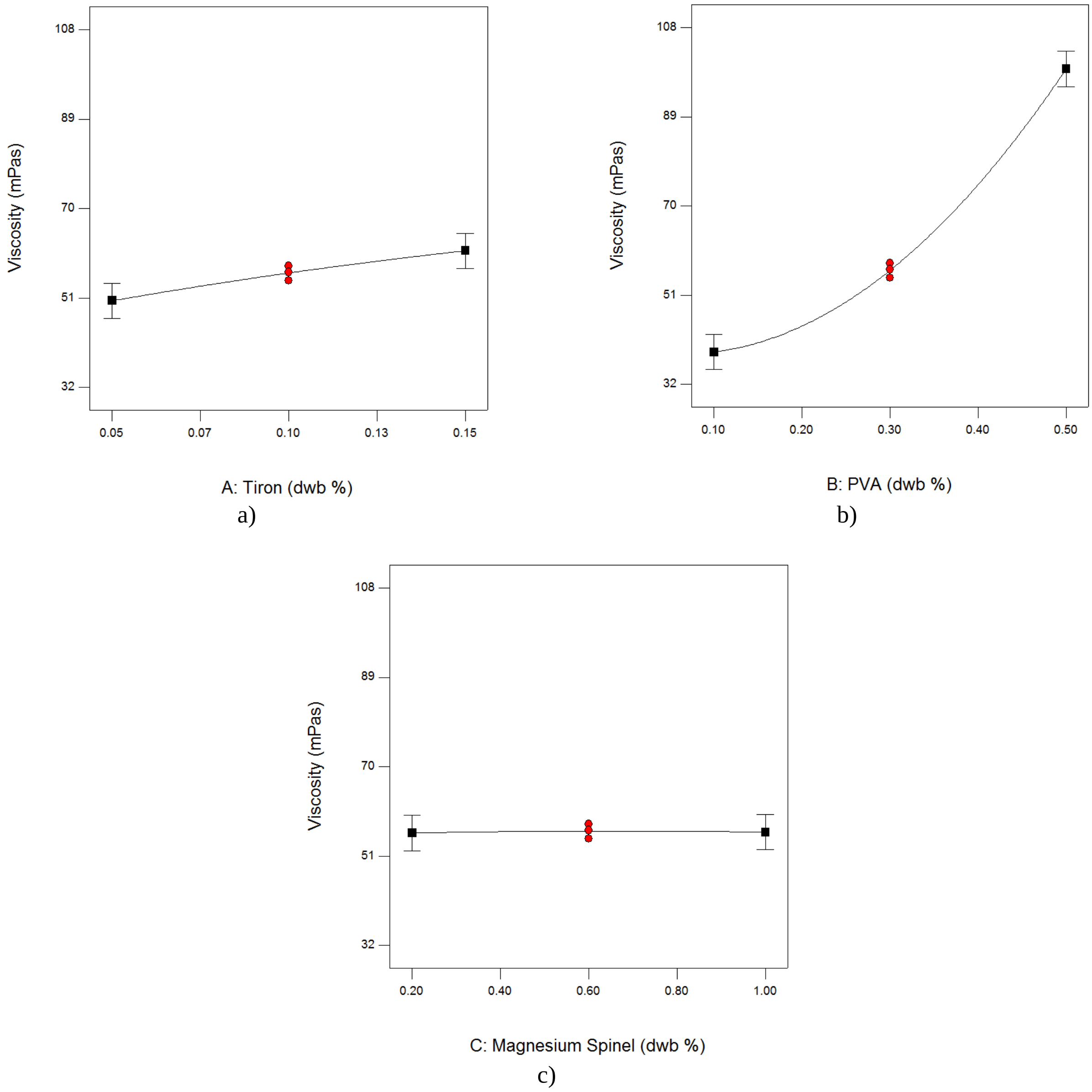

Fig. 2. a) Effect of Tiron onto viscosity, b) Effect of with PVA onto viscosity, c) Effect magnesium spinel onto viscosity

5. Conclusion

Manufacturing companies aim to increase profits by reducing manufacturing cost and environment impact of product by finding a more sustainable approach to product design and manufacturing. With that goal in mind, the reuse of recovered waste (secondary) alumina powder which was obtained after green machining in factory production of ceramics was investigated. Three independent variables considered for the preparation of stable highly concentrated alumina suspension which contain 20 dwb. % waste alumina powder were successfully optimized by the Box-Behnken response surface methodology. The minimum viscosity was determined by numerical optimization using Design Expert software. The viscosity was used for the suspension stability estimation. The optimum parameters for minimum viscosity (32.43 mPas) of investigated suspension's composition were achieved with the addition of 0.05 % Tiron, 0.1 % PVA and 0.2 % magnesium spinel expressed on dry weight basis. The model graphs suggest that the addition of PVA had a significant effect on viscosity (p<0.0001) whereas for the addition of Tiron and magnesium spinel (p>0.0001) suggested relatively lesser effect on viscosity of prepared alumina suspensions.

The study has shown that is possible to recycle the waste alumina powder which is generated during green machining of industrial production of technical ceramics. The obtained results also suggest that the waste alumina powder may be used to prepare new ceramic products by slip casting forming technique and eventually reduce manufacturing costs. The study was limited on preparation of stable alumina suspensions, formed by slip casting ceramic forming technique with fixed ratio of waste alumina powder of 20 dwb. %. The next step is to take into consideration the quality of the obtained product. Therefore, further research will be focused on finding balance between low viscosity which is necessary for obtaining complex shapes by slip casting method and mechanical (functional) properties such as hardness and wear resistance.

6. Acknowledgments

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