Evaluation of Mechanical and Metallurgical properties of different composition of Zirconia toughened alumina

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Abstract

In this study an attempt has been made to investigate the densification behaviour, microstructure, and mechanical properties of yttria (2-3%) doped in Zirconia mixed with different percentage of alumina (85-95%). The developed composites are uniaxially compressed into rectangular bars having 4 mm width 50 mm length and 4 mm thickness. The green compact specimens are sintered at different temperature ranging between 1200°C to 1600°C to know the thermal effect on mechanical properties. The sintered specimens can be further grounded to make inserts. The results show that the addition of additives like Cr_20_3, MgO and CeO_2 accelerated the densification and toughness of the developed composite. This investigation exploits the Box Behnken design approach with an overall objective of optimizing the powder synthesis parameters. The design correlates the effect of sintering parameters with the microstructure, density and mechanical properties of zirconia toughened alumina. For each response parameter regression model has been developed and the output result showed the adequacy of models almost at all condition. Effect of each input parameter on output responses have been investigated using RSM. 2.5 mol % yttria doped with 90 Wt % alumina sintered at 1600°C has been observed as optimum with the highest desirability (98%) for the mechanical properties.

Keywords: Zirconia Toughened Alumina, Densification, Sintering, Box Behnken Design, ANOVA

1. INTRODUCTION

The parameter like fracture toughness, hardness and flexural strength plays a vital role to attend the desired performance especially when fatigue loads, precision fits etc. are of concern. Previous literatures also show that flexural strength, hardness and strength depends on material properties such as microstructure and bonding between the phases in a ceramic composite. Zirconia toughened alumina (ZTA) is one of the most widely used composite oxide structural ceramics [1]. Zirconia toughened alumina, typically consists of 85 to 95 percent alumina with a 5 to 15 percent zirconia concentration, which enhances the strength and toughness of the alumina. Stress induced transformation toughening of zirconia particles gives high strength of the composite. Stress induced transformation toughening results in uniform internal strain, which causes the zirconia structure to switch into another phase and fill the crack. Due of this phase switch, the volume of zirconia particles increases and creates stresses within the alumina structure. These stresses effectively heal the crack and block its further propagation. The added zirconia doubles the composite's strength and enhances toughness between two to four times. Previous experiments [2-4] on ZTA revels that composites of ZTA has excellent mechanical properties, such as high strength, hardness, abrasion resistance and toughness. Effect of temperature on the Young's modulus, flexural Strength, and fracture of yttria-stabilized zirconia has been carried out by Admes [5]. The investigation illustrate that when the temperature increases, elastic moduli rapidly decreases up to a temperature of 1100ºC thereafter, a gradual decrease has been noted upto 1350ºC. Furthermore, various experiments on ZrO_2 were carried out by Abden et al. [6] which revealed that due to presence of ZrO_2 grains as inter- and intra-granular particles in the Al_2O_3 matrix grains, the composite 3YSZ - 40 wt% Al_2O_3 have highly dense microstructure with flexural strength of 340 MPa and Vickers hardness of 14.31 GPa. Pressure-less sintering of ZrO_2 reinforced with Al_2O_3 particles (ATZ) and Al_2O_3 reinforced with ZrO_2 particles (ZTA) was studied by Nevarez-Rascon et al. [7]. None of the previous works studied, the effect of simultaneous variation of mol percentage of yttria, alumina and sintering temperature, on the mechanical properties of Y-ZTA.

The purpose of this work is to develop regression models to predict the hardness, fracture toughness and flexural strength for yttria doped zirconia toughened alumina (Y-ZTA) developed by powder metallurgy routs. Models are developed by using response surface methodology (RSM) technique for response like hardness, fracture toughness and flexural strength. The aforesaid models then were utilized to predict the hardness, fracture toughness and flexural strength for different composition and sintering temperature. Analysis of variance (ANOVA) has been used to assess the contribution of process parameters like mol % of yttria, weight % of alumina and sintering temperature on hardness, fracture toughness and flexural strength of the composites.

2. EXPERIMENTAL PROCEDURE

2.1 Preparation of powder:

In this investigation Y_2O_3 (2, 2.5 and 3 mol %), stabilized ZrO_2 is prepared by mixing zirconium oxychloride octahydrate [$ZrOCl_2.8H_2O$] (Loba Chemie, Mumbai, India), and yttrium nitrate hexahydrate [$Y(NO_3)_3.6H_2O$] (Loba Chemie, Mumbai, India). The prepared YSZ powder is further mixed with α-Al_2O_3 (Merck, 99.85% purity, average particle size ~150 nm). Nine different compositions were prepared by varying the Al_2O_3 content as 85, 90 and 95 wt %, in all the possible combination with 2, 2.5 and 3mol % YSZ content. The developed powders were placed in a ball-mill for 12 hrs with 0.8% polyethylene glycol, using high purity alumina balls of 10 mm diameter. After ball milling the composites were dried in a oven at 100°C for 24 hrs. Then powder was gently crushed in a mortar pestle to get a homogenious mixed composite. In the end, the devolped powder is placed in a high temperature furnace to calcine at 800°C.

2.2. Preparation of sample:

The calcined powder is mixed with 5 wt% PVA (polyvinyl alcohol) solution for 60 min in ultrasonic machine followed by automatic stirrer for 30 min. The mixed wet powder is placed at

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100°C in oven for 6 hr to eliminate the water content of the binder solution. Green compaction of dried powder is done in a hydraulic press (Make: Carver, USA), with a die-punch arrangement as portrayed in Fig. 1, at a pressure of 5 ton cm^{-2}, to provide a rectangular shape (4 mm × 4mm × 50 mm) after sintering (Fig 2(a)). The green compacted samples are sintered at three different temperatures: 1200°C, 1400°C and 1600°C, with a dwell time of 2 hrs. The heating rate was maintained as 10°C/min up to 600°C; 8°C/min up to 1000°C; 4°C/min up to 1200°C; 3°C/min up to 1400°C; 2°C/min up to 1600°C. The cooling rate was 10°C/min until the inertia of the furnace prevailed.

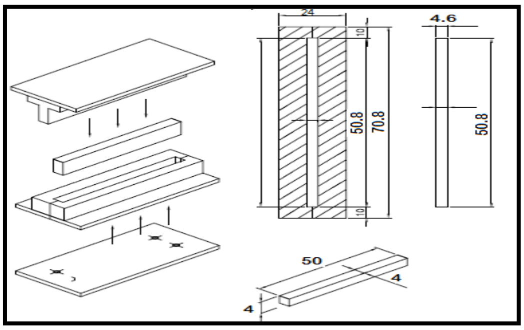

Fig. 1: Isometric and 2-D view of Die-Punch

2.3. Characterization of powders:

FESEM (CARL-ZEISS-SMT-LTD, Germany, Model: SUPRA 40) was used to know the microstructure of all prepared sample. The crystalline phases of the prepared composite specimens, sintered at 1600°C was studied by XRD-PW1710 with Cu-Kα radiation (λ = 0.15406 nm) in a continuous mode from 2θ range 20-70° at an angular step of 0.02° with a fixed counting time of 0.6 s/step. The voltage and current was kept at 40 kV and 40 mA, respectively. Flexural strength of each sample was determined using three point bending test on universal tensile machine as shown in Fig. 2 (b) (Tenius Olsen Serial No H50KS-0537) with span length set to 50 mm and the cross head displacement velocity kept at 0.5 mm/min.

The hardness and fracture toughness of each prepared sample was measured with the aid of a Vickers hardness testing machine (Matsuzwa, MXT-70) shown in Fig. 3. The indenter was used to make impression (usually no more than 0.5 mm) into the surface of the material at loads ranging up to approximately 2 Kgf shown in Fig. 3.

The Vickers number (HV) and fracture toughness K_{IC} is calculated using the following formula [8-9].

$$HV = 1.854(F/D^2) \quad (1)$$

$$K_{IC} = 0.16(c/a)^{-1.5}(Ha^{0.5}) \quad (2)$$

Where, F = applied load (Kgf), D^2 = area of the indentation (mm^2), K_{IC} = Fracture toughness ($MPa.m^{1/2}$), H = Vickers hardness (MPa), P = Test load (Newton), c = Average length of the cracks obtained at the tips of the Vickers marks (microns), a = Half average length of the diagonal (microns).

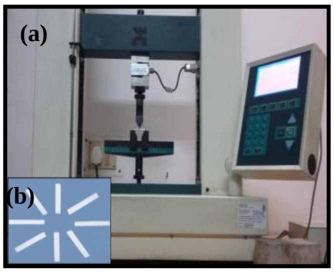

Fig. 2 (a) Universal Tensile Machine(Tenius Olsen Serial No H50KS-0537) (b) Prepared samples

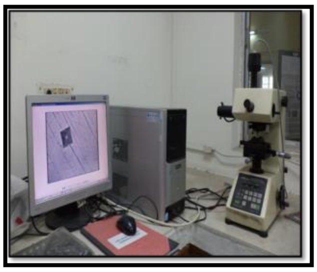

Fig. 3 Vickers hardness testing machine Matsuzwa, MXT-70

2.4. Box Behnken Design:

Box and Behnken experimental design for RSM, having three level designs for fitting response surfaces, were used. The design follows spherical design with all points lying on a sphere of radius $2^{1/2}$ [10], which are formed by combining 2^k factorials with incomplete block designs. Table 1 shows the design matrix generated for the Box-Behnken study (17 experiments). The statistical significance of Box-Behnken designs has been estimated through Analysis of Variance (ANOVA) [10]. The models are run up to their first order interaction terms. A linear regression partition generated by ANOVA, consisting total variation of a sample into components, is used to compute F-ratio that evaluates the effectiveness of the parameters. Each experiments are replicated three times to reduce type 1 error and increase the power of analysis.

Table 1: Experimental data of Box Behnken Design

S. NO	MOL% OF YTTRIA IN YSZ	WT % OF ALUMINA IN ZTA	SINTERING TEMPERATURE	HARDNESS (GPA)	FRACTURE TOUGHNESS (MPA $M^{1/2}$)	FLEXURAL STRENGTH (MPA)
1	2.5	85	1600	15.34	5.31	586.2
2	2.5	90	1400	16.32	5.85	587.1
3	2.0	85	1400	14.08	4.85	525.1
4	2.5	90	1400	16.44	5.77	595.3
5	2.5	90	1400	16.41	5.72	583.6
9	3.0	90	1600	16.54	5.62	598.3
7	2.5	95	1200	14.58	5.48	540.4
8	2.5	95	1600	16.25	6.02	615.1
9	2.5	85	1200	13.77	4.48	534.7
10	3.0	90	1200	14.85	5.30	532.1
11	2.5	90	1400	16.36	5.93	593.1
12	2.0	95	1400	15.03	5.27	563.2
13	2.0	90	1600	15.37	5.35	589.7
14	3.0	85	1400	14.84	5.43	569.1
15	2.0	90	1200	14.67	5.18	489.2
16	2.5	90	1400	16.38	5.86	591.2
17	3.0	95	1400	15.88	5.51	576.3

3. RESULT AND DISCUSSION

3.1. Characterization of prepared powder:

The analysis of phase composition, density, crystallite size, and grain size of different composites of ZTA shows that as the percentage of yttria and alumina increases the density of each sample increases. However, due to toughening mechanism, the grain size decreases which is in agreement with Mondal et.al. [1]. A typical X-ray Diffractometry of 2.5% YSZ Powders in 90% alumina is portrayed in Fig. 4. As expected from the phase diagram of the systems when yttria percentage is increased from 2 to 3 mol % the tetragonal phases of ZrO_2 gradually increased [1].

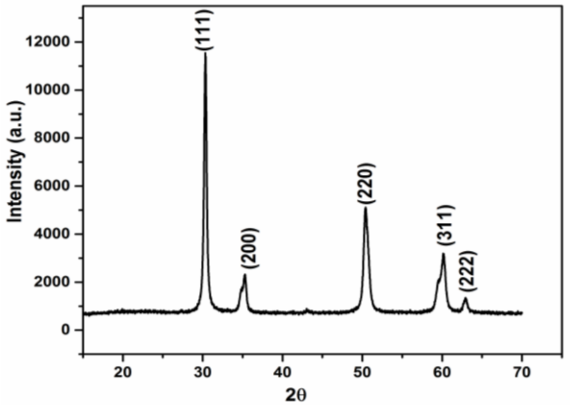

Fig. 4: A typical XRD of 2.5% YSZ

From Fig. 5 the FESEM images clearly show that zirconia grains are homogeneously dispersed throughout the alumina matrix. The excellent dispersion with homogeneous mixing is clearly observed in 2.5 mol % YSZ composite. It is also noticed that the agglomeration and voids at grain boundary is minimum for 2.5 mol % YSZ with 90 wt % alumina. Grain growth with increasing zirconia content has been noticed. It is observed that the metastable tetragonal phase of ZrO_2 in α-Al_2O_3 matrices has an important role for enhancing the hardness, fracture toughness and flexural strength of composites. The hardness, fracture toughness and flexural strength of alumina–zirconia composites are directly linked with the morphology and distribution of zirconia particles, their grain size, shape, location in alumina matrix, orientation, porosity, size distribution as well as boundary constitution [9]. These are critically influenced by the processing methodology and additive content as stabilizer.

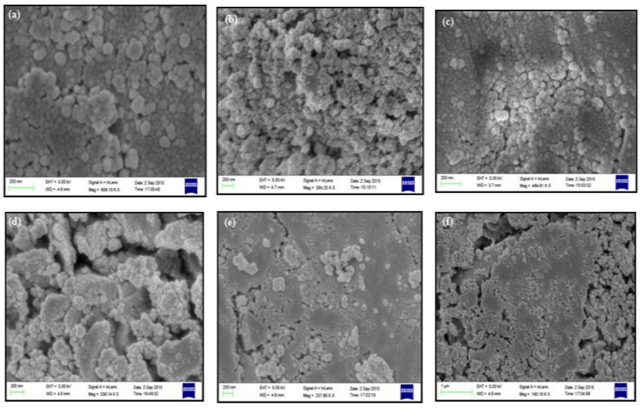

Fig 5 (a) FESEM image of 2 mol % YSZ (b) FESEM image of 2.5 mol % YSZ (c) FESEM image of 3 mol % YSZ (d) FESEM image of 2.5 mol % YSZ with 85 wt % Alumina (e) FESEM image of 2.5 mol % YSZ with wt 90% Alumina (f) FESEM image of 2.5 mol % YSZ with wt 95% Alumina.

Increase in hardness is attributed to the distribution of ZrO_2 grains as intergranular and intragranular particles in the Al_2O_3 grains [11]. Grain boundaries also work as an obstacle for plastic deformation and increases hardness [6]. It has been observed that the hardness of composite increases with increasing Al_2O_3 content. When the Wt % of yttria and alumina increases more than 2.5 Wt % and 90 wt% there is separation between the matrices, resulting in formation of pore or void inside the structure. Hence, the hardness starts decreasing as shown in Fig. 5. The fracture toughness of the prepared composites are more than that of conventional alumina (about 3.0 $MPa.m^{1/2}$). The highest hardness was obtained with 2.5 Wt % YSZ and 90 wt% alumina. This happens mainly due to the presence of metastable tetragonal ZrO_2, more than stable tetragonal/cubic phase, which in turn expands the volume and restrict the crack propagation. Therefore, it is assumed that both stress induced transformation in zirconia phase and residual stress in alumina phase are the predominant factors for higher fracture toughness of the newly developed composite. Maximum flexural strength was achieved with 2.5 mole % Y_2O_3.

3.2. Model Fitting and Statistical Analysis

Design Expert software (Version 8.0.1) has been used to estimate the affect of process parameter on hardness, fracture toughness and flexural strength. Furthermore, the experimental findings are used to develop a second-order polynomial regression equation with the inclusion of interaction terms. The adequacy of developed models is checked with 95% confidence level. The criteria to check the significance of each model is whether the F-ratio of the regression model is more than the critical value specified in the F-table for 95% confidence level. It is observed that all the three models portrayed in Table 2, Table 3 and Table 4 using quadratic equation satisfy the adequacy conditions in non-linear form. To remove the insignificant terms, backward elimination procedure has been applied.

Table 2: ANOVA for Hardness

Source	Sum of Squares	df	Mean	F Value	p-value Prob > F	
Model	13.222	7	1.89	140.642	< 0.0001	significant
A-Yttria (mol %) in YSZ	1.095	1	1.09	81.542	< 0.0001	
B-Al_2O_3 in ZTA	1.720	1	1.72	128.099	< 0.0001	
C-Sintering Temperature	3.962	1	3.96	294.995	< 0.0001	
AC	0.245	1	0.24	18.243	0.0021	
A^2	1.165	1	1.16	86.735	< 0.0001	
B^2	3.399	1	3.39	253.081	< 0.0001	
C^2	1.046	1	1.04	77.903	< 0.0001	
Residual	0.120	9	0.01			
Lack of Fit	0.112	5	0.02	10.604	0.0200	significant
Pure Error	0.008	4	0.002			
Cor Total	13.344	16				

From Table 2 it is clear that the developed model is significant. The table also shows that the sintering temperature mostly influences the hardness, as evident from its highest percentage square contribution (32%). It is followed by square percentage of Al_2O_3 which is nearly 27%. The combined effect of percentage of yttria and sintering temperature has very less effect (~ 2% contribution). This is possibly due to the grain growth of YSZ being retarded and full densification of the composite occurring at high sintering temperature. However, when temperature is increased beyond a limit, an adverse phenomenon i.e. reduction of tetragonal phase and increase in the cubic phase formation in the zirconia matrix decreases the hardness. The mean square percentage of “Residual” value and “Lack of Fit” are just 1.3% and 2.24% respectively, while the mean square percentage of “Error” is just 2.12%. This signifies that the develop model can be fitted to navigate the design space.

From Table 3 the "Model F-value" has been calculated as 9.35, which signify that the model is significant. The table also suggest that, the effect of percentage of alumina is highest among all processing parameter with 30% followed by sintering temperature 21% with percentage of alumina^2 having 19%. The microstructure analysis suggest that most of the ZrO_2 grains are located at the triple junctions of the Al_2O_3 grains and the grain boundaries. Due to the strong self-diffusion of alumina particles within alumina matrix, the zirconia particles are isolated at: grain boundaries, intermediate space between bigger alumina grains and some isolated pores present in the grain boundaries [6]. The low values of "Residual" "Lack of Fit" and "Error" percentage suggest that the developed model is navigating the design space very well. From Table 4 "Model F-value" is 38.77 satisfies the significance of model. The develop table also signifies that there is noteworthy effect of sintering temperature on flexural strength. The dependence of flexural strength on sintering temperature may be due to the reduced grain size and uniform distributions of grain growth. Sintering at high temperature. reduces thermal stress and thus improves strength. It is also noteworthy that the particle size lower than the critical value do not affect the strength during fracture test, as stated in [12]. The interaction and direct effect of all processing parameter on the response is incorporated in the Fig 6 and 7. From Fig 6 (a) the analysis shows that, simultaneous effect of sintering temperature and Yttria (mol %) in YSZ has significant effect on the hardness mainly due to transformation mechanism described in section 3.1. The interaction effect of flexural strength is shown in Fig 6 (b) and (c) which illustrate that all the three parameters have significant effect on the flexural strength. The direct effect of different percentage of Al_2O_3, on the responses, keeping sintering temperature and mole percentage of yttria as constant is incorporated in the Fig 7(a).

Table 3: ANOVA for Fracture Toughness

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F	
Model	2.160	6	0.360	9.344	0.0013	significant
A-Yttria (mol %) in YSZ	0.183	1	0.183	4.749	0.0543	
B-Al2O3 in ZTA	0.610	1	0.610	15.841	0.0026	
C-Sintering Temperature	0.432	1	0.432	11.220	0.0074	
A^2	0.286	1	0.286	7.414	0.0215	
B^2	0.380	1	0.380	9.865	0.0105	
C^2	0.173	1	0.173	4.502	0.0598	
Residual	0.385	10	0.038			
Lack of Fit	0.358	6	0.059	8.877	0.0266	significant
Pure Error	0.027	4	0.007			
Cor Total	2.546	16				

Table 4: ANOVA for flexural Strength

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F	
Model	17305.43	8	2163.179	38.774	< 0.0001	significant
A-Yttria (mol %) in YSZ	1474.24	1	1474.245	26.425	0.0009	
B-Al_2O_3 in ZTA	798.00	1	798.001	14.303	0.0054	
C-Sintering Temperature	10723.80	1	10723.801	192.219	< 0.0001	
AB	238.70	1	238.702	4.278	0.0724	
AC	294.12	1	294.122	5.272	0.0508	
A^2	2466.87	1	2466.871	44.217	0.0002	
B^2	232.44	1	232.441	4.166	0.0755	
C^2	770.78	1	770.782	13.816	0.0059	
Residual	446.31	8	55.7893			
Lack of Fit	357.82	4	89.455	4.043	0.1023	not significant
Pure Error	88.49	4	22.123			
Cor Total	17751.74	16				

3.3 Interaction and Direct effect of process parameter:

The graph shows that, with increase in Al_2O_3 all the responses viz. hardness, fracture toughness and flexural strength, first increases and then decreases. The results are in agreement with Mondal et.al. [1] as explained in section 3.2. Similar explanation is also applicable to the trends in Fig 7 (b). Fig 7 (c) shows that when sintering temperature increases hardness and flexural strength increases. This may be due to elimination of voids and grain separation.

4. Conclusion

The different composition of yttria stabilized zirconia toughened alumina has been developed to study the hardness, fracture toughness and flexural strength. The results revel that for 2.5 mol % yttria in zirconia and 90% alumina, sintering at a temperature is 1600°C, have maximum hardness (16.75 GPa), fracture toughness (5.85 MPa.m$^{1/2}$) and flexural strength (613.2 Mpa). The results of ANOVA signify that the models fitted well to the experimental results.

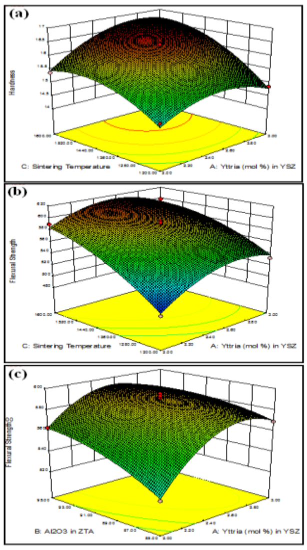

Figure 6 (a) Interaction effect of Hardness with Sintering Temperature and Yttria(mol%) in YSZ (b) Interaction effect of Flexural Strength with Sintering Temperature and Yttria(mol%) in YSZ (c) Interaction effect of Flexural Strength with percentage of Al_2O_3 and Yttria(mol%) in YSZ.

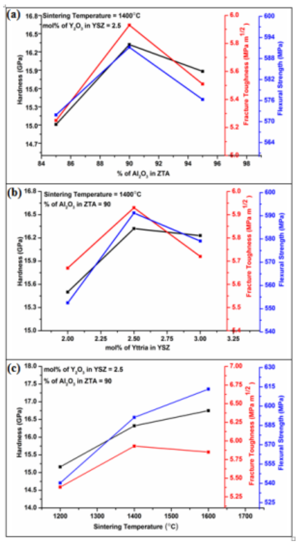

Figure 7 (a) Direct effect of Hardness, Fracture Toughness and Flexural Strength with mol% Yttria in YSZ (b) Direct effect of Hardness, Fracture Toughness and Flexural Strength with percentage of Al2O3 (c) Direct effect of Hardness, Fracture Toughness and Flexural Strength with Sintering Temperature.

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