Journal of Ceramic Processing Research. Vol. 11, No. 6, pp. 678~684 (2010)

J O U R N A L O F
Ceramic Processing Research

Relationship of the R-curve with the microstructure of alumina ceramics

Kuntal Maiti and Anjan Sil*

Department of Metallurgical and Materials Engineering, Indian Institute of Technology Roorkee, Roorkee-247 667, Uttarakhand, India

The influence of sintering temperature and soaking time period on R-curve behavior of Al_2O_3 ceramics have been investigated. Al_2O_3 samples were prepared by solid state sintering at 1500 °C, 1600 °C and 1700 °C for different soaking time periods of 3 h, 6 h, 9 h, 12 h, under an ambient atmosphere. However, longer soaking time periods of 18 h and 24 h were also applied for the samples sintered at 1500 °C. The fracture toughness of the sintered samples was determined from the cracks induced by the Vickers indentation method. The fracture toughness increases with the crack extension (R-curve behavior) for the samples having an average grain size limited to 7.3 μm. R-curve behavior was not obeyed for the samples having a grain size more than 7.3 μm. The R-curve nature has been predicted from the predominant intergranular crack extension mode as has been observed from microstructures of fracture surfaces of the samples. Evidence of crack deflection was found for the samples sintered at 1500 °C and 1600 °C for which relatively higher fracture toughnesses of 5.28 ± 0.08 MPam$^{1/2}$ and 5.37 ± 0.25 MPam$^{1/2}$ respectively, were determined. The samples sintered at 1700 °C have a relatively lower fracture toughness (4.86 ± 0.21 MPam$^{1/2}$). Transgranular fracture in the samples sintered at 1700 °C for a soaking time of 6 h and more containing larger grains is more pronounced. Evidence of crack deflection by studying the fracture surface and fracture surface roughness characteristics using Field Emission Scanning Electron Microscope (FESEM) and Atomic Force Microscope (AFM) respectively has been shown.

Key words: Sintering, Fracture toughness, R-curve, Vickers indentation methods.

Introduction

The increasing fracture resistance with crack extension in ceramics (R-curve behaviour) is highly desirable for their use in structural applications. In noncubic and nontransforming ceramics, the magnitude of this increase is quite appreciable, depending on the microstructure. The R-curve has an influence on crack growth and hence on flaw tolerance which is critical for ceramics used in structural design. This phenomenon has been explained by a grain-bridging model which was developed in 1989 by Bennison and Lawn [1]. Miyahara *et al.* have also explained the R-curve phenomenon with a grain bridging effect [2]. The grain bridging is a phenomenon, where interlocking grains along the crack wake partially shield the crack tip from the applied stress and the fracture resistance increases with crack extension due to the growing number of bridging grains. Deng *et al.* [3] reported the effect of residual stress on the R-curve behavior of particle reinforced ceramic composites. The residual stresses develop in the matrix of ceramic composites due to mismatch of thermal expansion coefficients between the different ceramic phases. Researchers already reported the R-curve behavior for Al_2O_3 ceramics as well as particle and whisker reinforced ceramic matrix composites [3]. The direct correlation between the roughness of the fracture surface, as a measure of the degree of the crack deflection, and the fracture toughness of the ceramic composites has been suggested by various researchers [4, 5]. Miyahara *et al.* reported that the fracture toughness increases with increasing grain size of sintered Al_2O_3 upto the size of 6 μm and intergranular fracture is the predominant mechanism for the enhancement of the fracture toughness [2]. However, different test methods such as chevron notch, bridge indentation, controlled surface flaw and fatigue pre cracking were used by the researchers. Tomaszewski *et al.* [6]. reported that the crack growth resistance of Al_2O_3 ceramics increases significantly with an increase of the Al_2O_3 grain size upto 67.4 μm and residual stress due to crystallographic and thermal expansion mismatch between adjacent grains during cooling is the toughening mechanism for that. They have also shown that a decrease in the grain size reduces the grain bridging as well as residual stress [6]. Faber and Evans [7] and Deng *et al.* [8]. reported that for the enhancement of the fracture toughness in ceramic composites which normally have smaller grains, crack deflection is responsible. They reported the second phase deflects the crack path. Tani *et al.* [9]. reported that the fracture toughness of Al_2O_3 ceramics decreases sharply with increasing grain size. Therefore the issue of microstructural dependence in view of the influence of grain size on the fracture toughness of Al_2O_3 ceramics has not been fully settled. In order to address this issue in the present study, Al_2O_3 ceramics have been prepared with different grain sizes which has been realized by sintering the ceramic powder compacts

*Corresponding author:
Tel : +91-1332-285073
Fax: +91-1332-285243(273560)
E-mail: anj_sil@yahoo.co.uk

at different temperatures (i.e. 1500 ºC, 1600 ºC and 1700 ºC) for different soaking time periods (i.e. 3, 6, 9. 12 h) at each temperature. The crack length variation due to indentation in each sample has been effected by applying indentation with different loads (i.e. 5, 10, 15, 20, 30 and 40 kg).

Experimental Procedure

The raw material used was Al_2O_3 (Industrial Ceramics, 99.8%, 0.4 μm) powder which was mixed with binder (polyvinyl alcohol) for granulation. The granulated powder was compacted in the form of rectangular bars of 3 × 5 × 45 mm^3 size, uniaxially in a hydraulic press at different loads between 15×10^3 and 22.5×10^3 kg. The green compacts were sintered at 1500 ºC for different soaking time periods of 3 h, 6 h, 9 h, 12 h, 18 h and 24 h. Samples were also prepared by sintering at 1600 ºC and 1700 ºC for the soaking time periods of 3 h, 6 h, 9 h and 12 h. The rate of heating as well as of cooling used in the sintering process was 10 K·$minute^{-1}$. The density of all samples was measured following Archimedes principle using water as an immersion medium. The samples were ground using silicon carbide (SiC) papers having mesh sizes of #120, #220, #320 to develop a smooth surface finish on one face. The smooth surfaces of the samples were polished with diamond pastes of 6 and 3 μm sizes, to the level of a mirror like finish, and indented by various loads of 5, 10, 15, 20, 30 and 40 kg for an identical dwell time of 15 s. The indentations were carried out under ambient conditions. After indentation, the length of each diagonal of the square shaped Vickers indentation and crack length generated from the indentation boundaries were measured by an optical microscope (Axiovert 200 MAT, ZEISS). The hardness (H_v) of each sample was calculated from the equation given below:

$$H_v = P/A = \alpha\ P/d_o^2 \quad (1)$$

where P is the applied load, A is the pyramidal contact area of the indentation, d_o is the average length of the diagonals of the resultant impression and $\alpha = 1.8544$ for Vickers indenter.

The measured lengths of the cracks for each indentation were averaged out and hence the fracture toughness (K_{IC}) was determined following the relationship given below [10]:

$$K_{IC} = \chi(E/H_v)^{1/2}P/a^{3/2} \quad (2)$$

where E is Young's modulus, H_v is the Vickers hardness, a is the radial crack length measured from the centre of the indent and χ is an empirical calibration constant = 0.016 ± 0.004.

The grain structure developed in the samples during sintering was seen from microstructural observations using FESEM (FEI, QUANTA 200F) and for this purpose the sample surface was polished and gold coated. The grain size of the sintered samples was measured using Image J software. Measurement of roughness of the fracture surfaces was carried out by AFM (NT-MDT Integra).

Results and Discussion

The different Al_2O_3 samples prepared by sintering at different temperatures and for various soaking periods are represented by $A_{t(hr)}{}^{T(^oC)}$ (e.g. $A_3{}^{1500}$ represents the pure Al_2O_3 sample sintered at 1500 ºC and soaked for 3 h).

Table 1 shows the fracture toughness and crack length in the samples sintered at 1500 ºC, 1600 ºC and 1700 ºC

Table 1. Average grain size, average crack length and fracture toughness of the samples sintered at 1500 ºC, 1600 ºC and 1700 ºC for the different soaking time periods

Sample	Avg. grain size (μm)	Average crack length (μm) at different load						Fracture toughness ($MPam^{1/2}$)	Average fracture toughness ($MPam^{1/2}$)
		5 kg	10 kg	15 kg	20 kg	30 kg	40 kg		
$A_3{}^{1500}$	0.688	48.5	94.08	115.52	124.75	173.48	188.25	5.22 ± 0.39	
$A_6{}^{1500}$	0.837	47	81.25	121.5	135	181.25	222.5	5.15 ± 0.19	
$A_9{}^{1500}$	0.843	50.7	86.8	100.7	130.72	186.53	227.5	5.38 ± 0.46	5.28 ± 0.08
$A_{12}{}^{1500}$	0.939	53.25	80	113.75	123.3	154.45	218.5	5.37 ± 0.55	
$A_{18}{}^{1500}$	4.81	-	84.3	98.28	132.79	158.44	230.28	5.27 ± 0.23	
$A_{24}{}^{1500}$	4.93	-	78.05	129.94	146.72	197	230.01	5.28 ± 0.19	
$A_3{}^{1600}$	5.35	53.45	93.8	181.57	169.6	182.03	282.01	5.44 ± 0.36	
$A_6{}^{1600}$	5.42	45.97	82.53	116.03	137	184.89	229.21	5.45 ± 0.27	5.37 ± 0.25
$A_9{}^{1600}$	5.57	-	95.92	147.88	184.6	211.78	239	5.57 ± 0.52	
$A_{12}{}^{1600}$	6.07	-	100.68	145.76	160.6	216.5	253	5.01 ± 0.42	
$A_3{}^{1700}$	7.30	-	151	165.86	180	191.55	261.88	4.56 ± 0.72	
$A_6{}^{1700}$	13.75	-	89.7	107.15	144.63	251.36	301.43	4.98 ± 0.49	4.86 ± 0.21
$A_9{}^{1700}$	15.46	-	94.48	122.45	199.27	286.4	299.51	4.85 ± 0.27	
$A_{12}{}^{1700}$	17.11	55.63	82.8	166.27	257.23	289.39	305.6	5.03 ± 0.49	

for different soaking time periods. The fracture toughness of the samples sintered at 1500 °C varies from 5.15 to 5.38 $MPam^{1/2}$ and the maximum crack length observed was 230.28 μm for the sample sintered for 18 h and the indentation load was 40 kg. The samples sintered at 1600 °C show marginally higher average fracture toughness, lying within the range of 5.01-5.57 $MPam^{1/2}$ and a maximum crack length of 282.01 μm has been observed for the soaking time of 3 h. Fracture toughness of the samples sintered at 1700 °C is lower, lying in the range of 4.56-5.03 $MPam^{1/2}$ and has a maximum crack length of 305.6 μm in the sample soaked for 12 h.

Microstructural analysis

Crack deflection path

Crack paths shown in Fig. 1(a)-(b) reveal that the crack propagation is mainly of an intergranular nature showing the crack deflection is the predominant mechanism for toughness enhancement.

The micrograph (Fig. 2) shows the transgrannular fracture of an Al_2O_3 sample sintered at 1700 °C for 3 h. Transgrannular fracture is the consequence to lower fracture toughness in the high temperature(1700 °C) sintered samples.

R-curve measurements

In Fig. 3, the measured K_{IC} values are plotted as a function of the crack size for Al_2O_3 ceramics sintered at 1500 °C. The variation in the fracture toughness obtained from several indentations for a given load has been presented by showing an error bar. The linear fit of the data points shows an increase in K_{IC} with an increase in the crack length extension, showing the R-curve behavior of the samples. The linear fit of the average K_{IC} s for each load, shows the increase in K_{IC} with crack length extension. However, the increase is lower for the samples sintered for 18 h and 24 h compared to that 3, 9 and 12 h.

From the Fig. 4 one can say that all the samples sintered at 1600 °C follow the R-curve behavior because a linear fit of the data points in the graphs shows increases in K_{IC} with increases in crack length. Crack deflection is the reason to maintain the R-curve in the samples sintered at 1500 °C and 1600 °C where grain bridging is not the main mechanism behind that for a smaller grain size [6]. The linear fit of the average K_{IC} values shows a continuous increase in K_{IC} with the crack length extension. Therefore the samples sintered at 1600 °C obey R-curve behavior.

The increase in fracture toughness with increase in the crack length extension has been observed for the sample sintered at 1700 °C for 3 h. The samples sintered at 1700 °C for 6 h, 9 h and 12 h show decrease in fracture toughness although marginally with the crack length extension (see Fig. 5).

Fracture surfaces of the samples sintered at 1500 °C, 1600 °C and 1700 °C are shown in Fig. 6. In Fig. 6(a) the presence of smaller grains is clear and the surface topo-

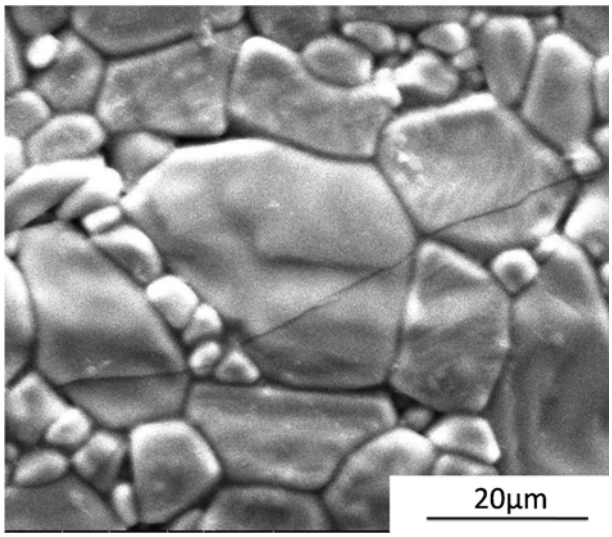

Fig. 2. SEM micrograph of an Al_2O_3 sample sintered at 1700 °C for 3 h.

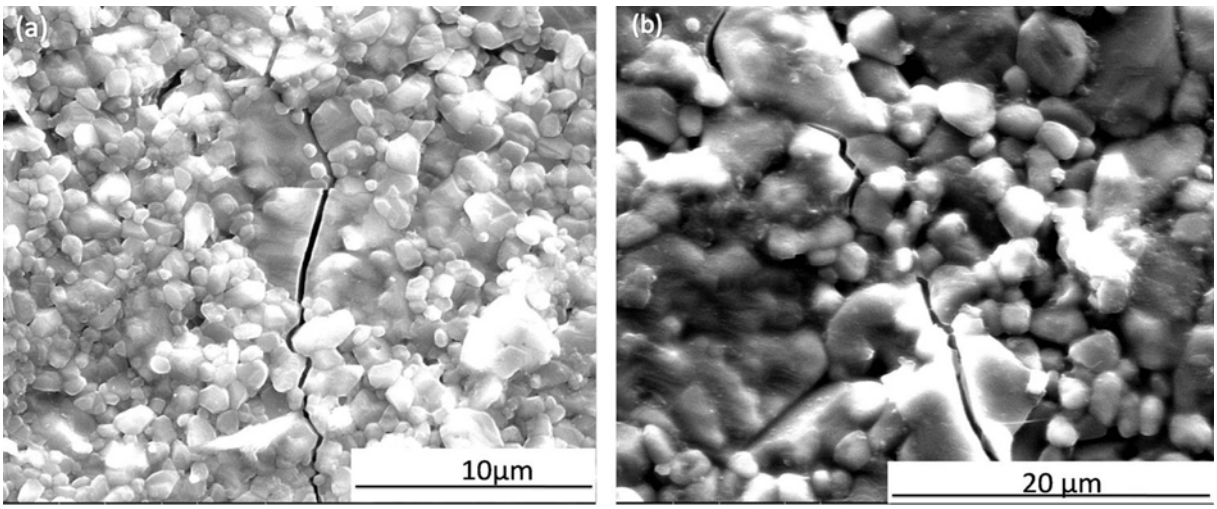

Fig. 1. SEM micrographs of Al_2O_3 sample sintered at (a) 1500 °C for 3 h (b) 1600 °C for 3 h.

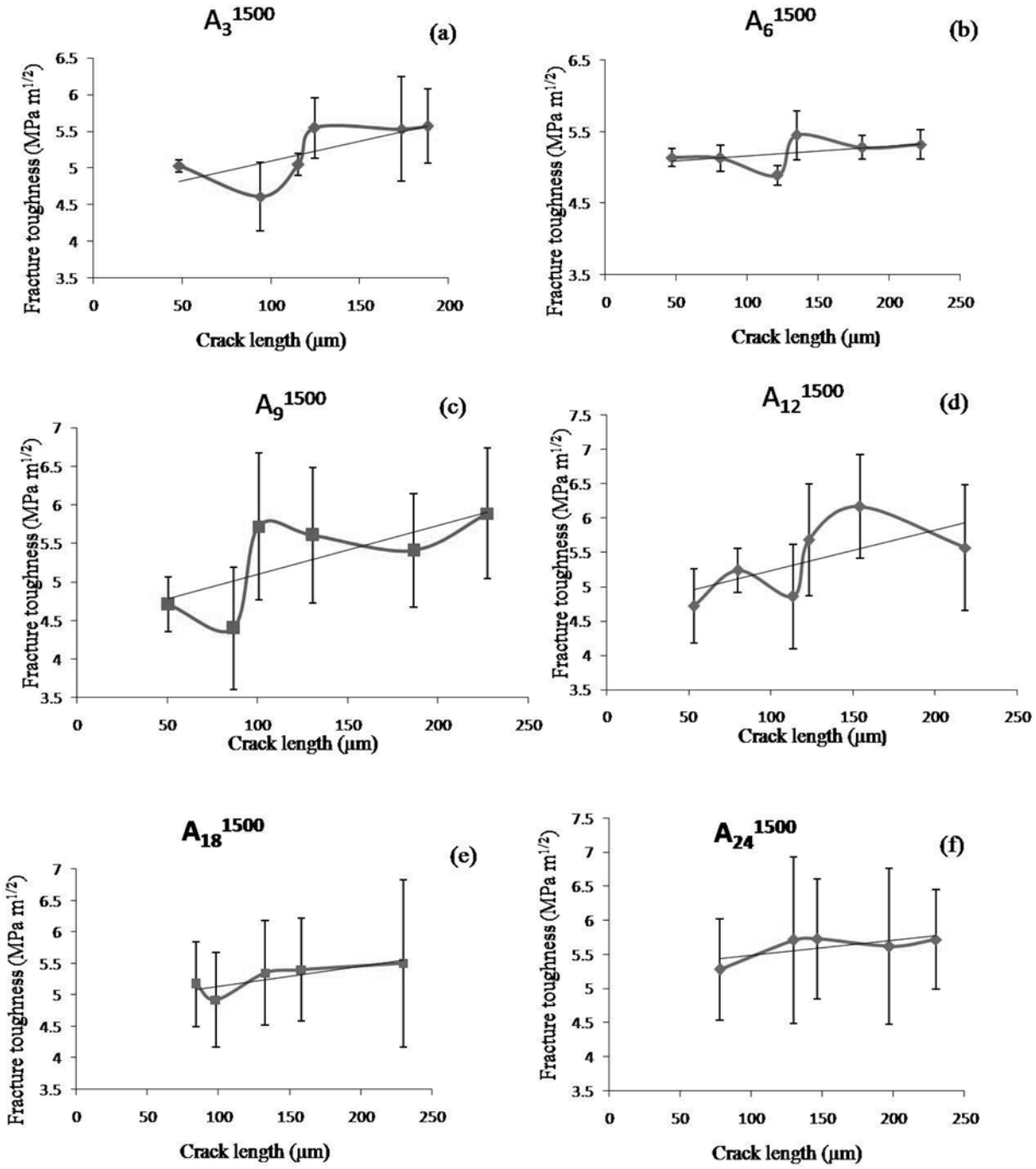

Fig. 3. Crack extension resistance of Al_2O_3 ceramics sintered at 1500 ºC for (a) 3 h, (b) 6 h, (c) 9 h, (d) 12 h, (e) 18 h and (f) 24 h.

graphy shows that the fracture has taken place predominantly by the intergranular mode. It is also clear from Fig. 6(b) that the smaller grains remain in almost the same size range as obtained in the case of Fig. 6(a), however the larger grains have grown substantially in relation to those in Fig. 6(a). Further the fracture surface topography confirms that fracture has taken place due to a mixed mode of intergranular and transgranular fracture, with a larger proportion of the intergranular mode. The fracture toughnesses of these samples (Fig. 6(b), (c)) show relatively higher toughness as listed in Table 1. But in Fig. 6(d) the flat surface topography over a considerable region in the micrograph area shows that the fracture has occurred predominantly due to a transgranular mode. The lower fracture toughness obtained is the obvious consequence of such a fracture surface feature. Fig. 6(c) shows that a large portion of the grain structure area is occupied by large grains with a smaller area portion filled with smaller sized grains. Although fracture of a transgranular type is present, but an intergranular fracture mode is clear from the Fig. 6(c)

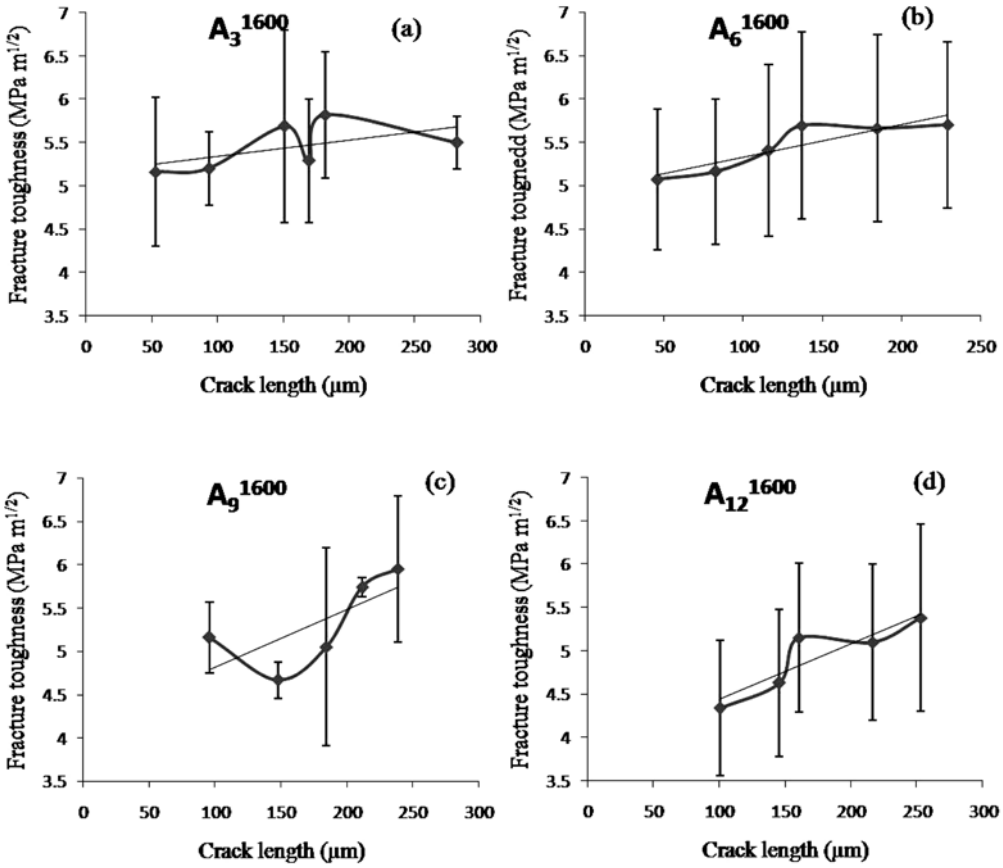

Fig. 4. Crack extension resistance of Al_2O_3 ceramics sintered at 1600 ºC for (a) 3 h, (b) 6 h, (c) 9 h and (d) 12 h.

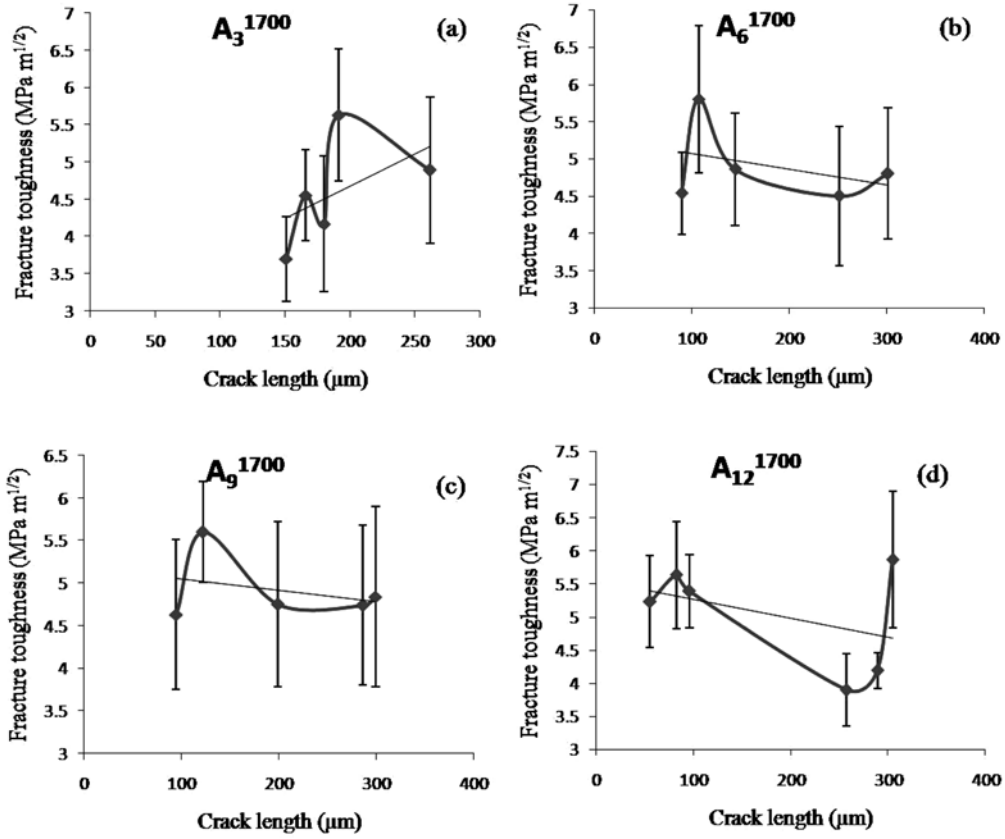

Fig. 5. Crack extension resistance of Al_2O_3 ceramics sintered at 1700°C for (a) 3h, (b) 6h, (c) 9h and (d) 12h.

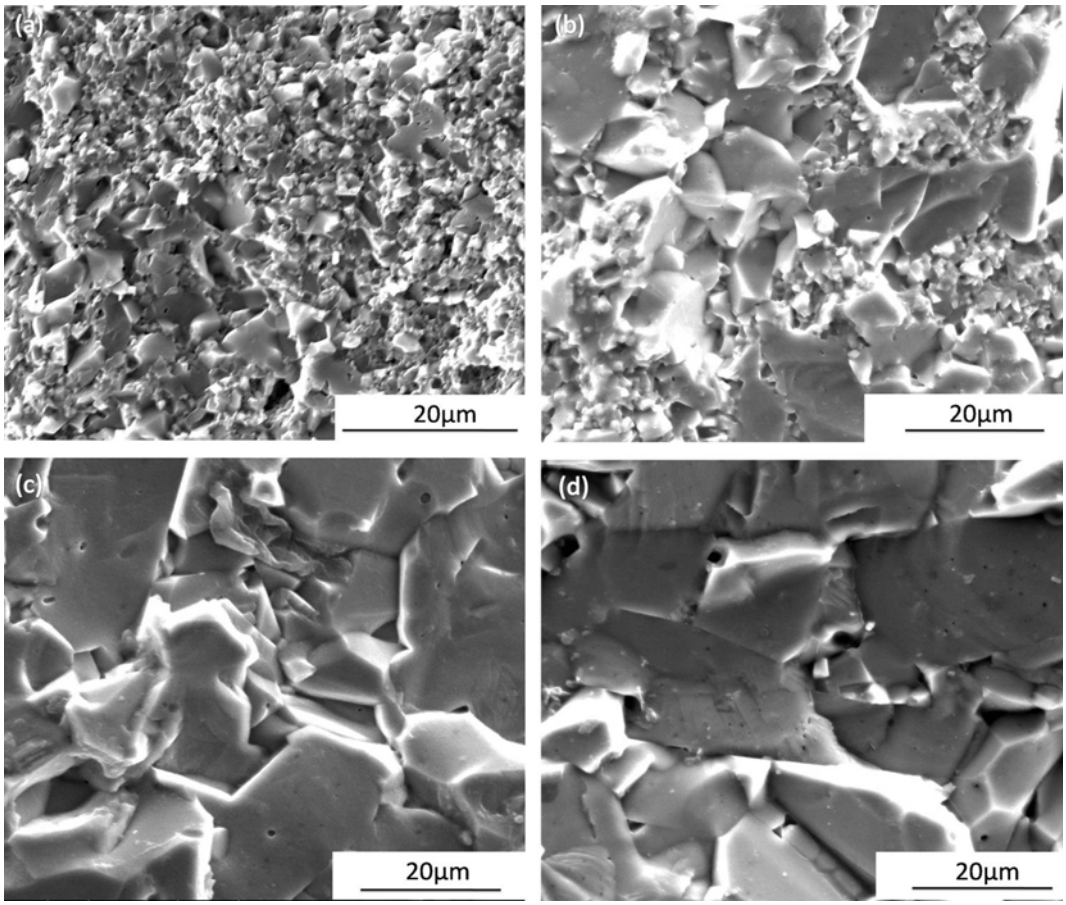

Fig. 6. SEM micrographs of fracture surfaces of Al_2O_3 sample sintered at (a) 1500 °C for 3 h, (b) 1500 °C for 18 h, (c) 1600 °C for 3 h and (d) 1700 °C for 3h.

and also from Fig. 1(b).

Fig. 7 shows data points plotted between fracture toughness and fracture surface roughness of the samples sintered at 1500 °C, 1600 °C and 1700 °C. A best fit line has been drawn through these data points. It can be seen that data points in the higher range of fracture toughness fall on or very close to the straight line whereas, the points in the lower fracture toughness region are situated far from the straight line. This means that the samples with a lower fracture toughness do not follow the same linear relationship when extended to the lower fracture toughness range. Boccaccini and Winkler have shown a linear correlation between the fracture toughness and fracture surface roughness which gives rise to crack deflection as the toughening mechanism in a Al_2O_3 platelet reinforced glass matrix composites [4].

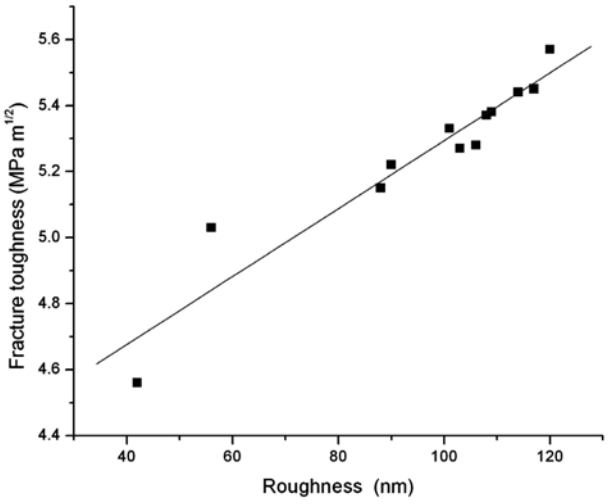

Fig. 7. Fracture toughness vs fracture surface roughness of the samples sintered at 1500 °C, 1600 °C and 1700 °C.

Conclusions

The following are the conclusions of the present study:

The Al_2O_3 ceramics prepared by solid state sintering at three different temperatures of 1500 °C, 1600 °C and 1700 °C for different soaking times have an average grain size variation from as low as 0.688 μm for 1500 °C sintered for 3 h to as high as 17.11 μm for 1700 °C sintered for 12 h.

The sintered samples having a grain size upto 7.3 μm show R-curve behavior. For the samples having an average grain size above 7.3 μm, the crack length extension does not follow the R-curve nature.

A fracture surface study reveals that a mixed mode of fracture i.e. intergranular and transgranular types with a larger proportion of former type cause fracture in the samples sintered at 1500 °C and 1600 °C. Such an observation leads to the conclusion that crack deflection is the primary reason for higher fracture toughness in these samples.

Transgranular fracture as a dominant mode of fracture for the samples sintered at 1700 °C is evident from the fracture surface study.

A linear relationship between the fracture surface roughness and the fracture toughness is well represented for the samples having higher fracture toughness.

References

1. S.J. Bennison and B.R. Lawn, Acta Mater. 37 (1989) 2659-2671.
2. N. Miyahara, H. Yamaishi, Y. Mutoh, K. Uematsu and M. Inoue, J. Jap. Soc. Mech. Eng. 37 (1994) 231-237.
3. Z.Y. Deng, J.L. Shi, Y. Shi, D.Y. Jiang and J.K. Guo, J. Mat. Sci. Lett. 19 (1997) 977-981.
4. A.R. Boccaccini and V. Winkler, Composites:Part A 33 (2002) 125-131.
5. K.P. Gadkaree, J. Mater. Sci. 26 (1991) 4845-4854.
6. H. Tomaszewski, M. Boniecki and H. Weglarz, J. Eur. Ceram. Soc. 20 (2000) 2569-2574.
7. K.T. Faber and A.G. Evans, Acta. Metall. 31 (1983) 577-584.
8. Z.Y. Deng, Y. Zhou, M.E. Brito, J.F. Yang and T. Ohji, J. Am. Ceram. Soc. 86 (2003)1789-1792.
9. T. Tani, Y. Miyamoto and M. Koizumi, Ceramics International 12 (1986) 33-37.
10. G.R. Anstis, P. Chantikul, B.R. Lawn and D.B. Marshall, J. Am. Ceram. Soc. 64 (1981) 533-538.