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Effect of grain size and residual stresses on R-curve behaviour of alumina based composites

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Abstract

Alumina samples with average grain sizes ranging from 2.6 to 67.4 μm and alumina based composites containing 10–30 wt.% of second phase particles with lower (SiC) and higher thermal expansion coefficient ($ZrO_2 + 8$ mol% Y_2O_3) were prepared. The effect of grain size and residual stresses on R-curve behaviour of ceramics was investigated by testing controlled crack growth during three point bending of single-edge-notched samples. The tests showed that R-curve behaviour of alumina ceramics strongly decreases with the decrease of Al_2O_3 grain size. Observed changes were related to the amount of bridging grains on the path of the propagating crack and residual stresses present in ceramics. The same tests done on composite samples showed that ZrO_2 and SiC addition also strongly decrease the effect of R-curve in alumina matrix. This phenomenon was explained by analysis of Al_2O_3 grain size, amount of bridging grains and residual stresses in ceramics.

Keywords: Al_2O_3; Al_2O_3–SiC; Al_2O_3–ZrO_2; Crack growth; Grain size; Residual stress; R-curve

1. Introduction

Normally the toughness-curve (T- or R-curve) behaviour is characterised by a change, usually showing an increase in toughness due to a material's resistance to crack propagation with crack extension. In many brittle solids this increase originates from crack tip shielding mechanisms that operate in the crack wake, exerting a crack closure force that decreases the net stress intensity at the crack tip. The R-curve results from the accumulation of this closure force with crack growth, typically reaching a saturation limit. It is well established that the R-curve behaviour of alumina is due to the formation of frictional traction (grain bridges) between opposing crack faces in the crack wake [1]. Microstructure variables that are known to control the level of toughening achievable via this mechanism include grain size and shape as well as the magnitude of internal stresses [2] originating from the combination of elastic modulus and thermal mismatch. It is generally known that residual stresses may be enhanced by the addition of the second phase, whose thermal expansion coefficient shows the desired degree of mismatch with that of the matrix. However, up to now, it was not explained if these additional fields of stress influence the amount of bridging grains or the value of frictional forces acting during grain pull out of the embedding matrix, only.

To solve this problem the effect of SiC and ZrO_2 addition (a phase with lower: $\alpha_{SiC} = 3.5–4.2 \times 10^{-6}$ °C^{-1}, $\alpha_{Al_2O_3} = 9.0 \times 10^{-6}$ °C^{-1} and higher thermal expansion coefficient: $\alpha_{ZrO_2} = 12 \times 10^{-6}$ °C^{-1}) on residual stress state change in alumina matrix, amount of bridging grains and the resulting altered crack behaviour were investigated in the present study.

2. Experimental methods

Alumina ceramics of the following chemical composition: Al_2O_3–99.55 wt.%, MgO–0.20 wt.%, Y_2O_3–0.25 wt.% was used as a matrix. Alumina powder was high purity (4 N concentration) with an average grain size below 0.5 μm. The starting SiC powder was β-phase with an average particle size about 1 μm. Matrix powder was mixed with SiC (0–30 wt.%) in ethanol and

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then hot-pressed in carbon dies lined with BN layers under a pressure of 15 MPa and a temperature of 1700°C for 1 h in an argon atmosphere. To the second type of mixtures a $ZrO_2 + 8$ mol% Y_2O_3 powder (MSY8, Mandoval Ltd) with an average grain size of 0.4 μm was used. In this case alumina powder mixed with ZrO_2 (0–30 wt.%) in distilled water was cold pressed and then sintered at 1700°C for 1.5 h in high vacuum. Sintered specimens were cut and ground yielding bars having the dimensions of 5×5×45 mm (square) or 1.5×6×45 mm (rectangular), and one surface was polished. In the central part of the rectangular bar sample (1.5×6×40) a sharp notch was prepared for testing the crack propagation behaviour. The polished surface of the samples was covered with a 150 nm thick Al layer to improve the crack path visibility during the test.

To explain the effect of matrix grain size on R-curve behaviour, the alumina samples with average grain sizes ranging from 2.6 to 67.4 μm were prepared by sintering at 1500–1900°C for 1–20 h in a vacuum furnace.

The tests of controlled crack growth were performed in three-point bending with 1 μm/min loading speed and 40 mm bearing distance using a universal testing machine (Model 1446, Zwick). The crack was initiated and slowly grew up by repeated loading and removing the load. The crack growth was controlled by observation of a stress-deflection curve. The load was removed when deflection of the sample increased at constant or decreasing stress. This procedure resulted in less than 100 μm increase of crack length by one step. The crack length c, was measured in situ using a CCD camera coupled to an appropriate microscope, which was fitted to the test equipment by a system of elevator stages driven by stepping motors. This enabled the precise movement of the microscope objective in x-y-z directions for adjustment, focussing and tracking on the beam side and bottom surface where the crack propagated. A measuring and registration system (framegrabber) was coupled with the load and strain system of the testing machine. Both systems were computer controlled. The optical and electronic magnifications were about 250×. The stress intensity factor K_I was calculated from the crack length c, and force P.[3] The data of $K_I = f(c)$ obtained in the range of crack length studied were fitted by a linear function $y = ax + b$ and the slope a, was used as a parameter describing R-curve behaviour. The linear dependence $K_I = f(c)$ obtained (especially for coarse-grained alumina) is in contradiction to the literature.[7] All experiments from literature show first a strongly increasing R-curve and then, depending on the initial crack size, a more flat part. It is well known from cyclic fatigue experiments that the most bridging interactions are destroyed after a few cycles.[8,9] In the present paper the crack increments between unloading are about 100 μm. resulting in more than 30 cycles for total crack extension. This degradation of bridging interactions must lead to a moderate increase of the R-curve. Consequently, the results obtained are not the R-curves in the common sense, but more "cyclic" R-curves. All experiments were done at room temperature in normal air environments.

The path of the crack was registered by SEM using OPTON DM950 microscope. Further computer analysis of the path resulted in an amount of bridging grain as a function of Al_2O_3 grain size obtained as a part (%) of a whole path length of the crack propagating through an alumina ceramics and which surrounds a bridging grains. The same values were calculated for alumina based composites.

Residual stresses within the alumina were measured using the piezospectroscopic technique described elsewhere.[4] An optical microscope was used to both excite the fluorescence and to collect and analyse the resulting fluorescence spectrum using an attached spectrometer (DILOR X4800). The 514.5 nm line of an argon ion laser was used to excite the fluorescence. The fluorescence signals were collected from a region of about 50 μm diameter in size. The intensity of the R_1 and R_2 fluorescence lines was scanned by integrating over 0.5 s intervals at a spacing of 0.2 wavenumbers with the intensity being recorded under computer control. The collected data were subsequently analysed with curve-fitting algorithms (double Lorenz function). The line position was identified by simultaneously fitting the R_1 and R_2 peaks using the NiceFit software package. All measurements were performed at room temperature. The peak shift due to temperature fluctuation was corrected using the ruby calibration. The instrumental shift was also corrected by simultaneously monitoring a characteristic Neon line at 14564 cm^{-1}. The average residual stress in the alumina matrix was calculated from the measured frequency shifts according to a relation of linear proportionality through the average piezospectroscopic coefficients given by He and Clarke[5].

Microstructure observations of ceramics studied were performed on polished and thermally etched surfaces by SEM using OPTON DSM 950 microscope. Grain size distribution measurements have also been made.

3. Results and discussion

As mentioned by Bennison et al.,[1] due to anisotropy in crystallographic and thermal properties of alumina, some grains in the alumina matrix are subjected to compression and play the role of "bridges". The remaining grains, subject to tension, are considered as making up the constitutive "matrix". The bridging grains, wedged in the microstructure by this internal compressive stress, lead to an increase in fracture toughness as the crack grows.

Such bridging grains, surrounded by a propagating crack were observed in samples studied during controlled crack growth tests (see Fig. 1). A dependence of

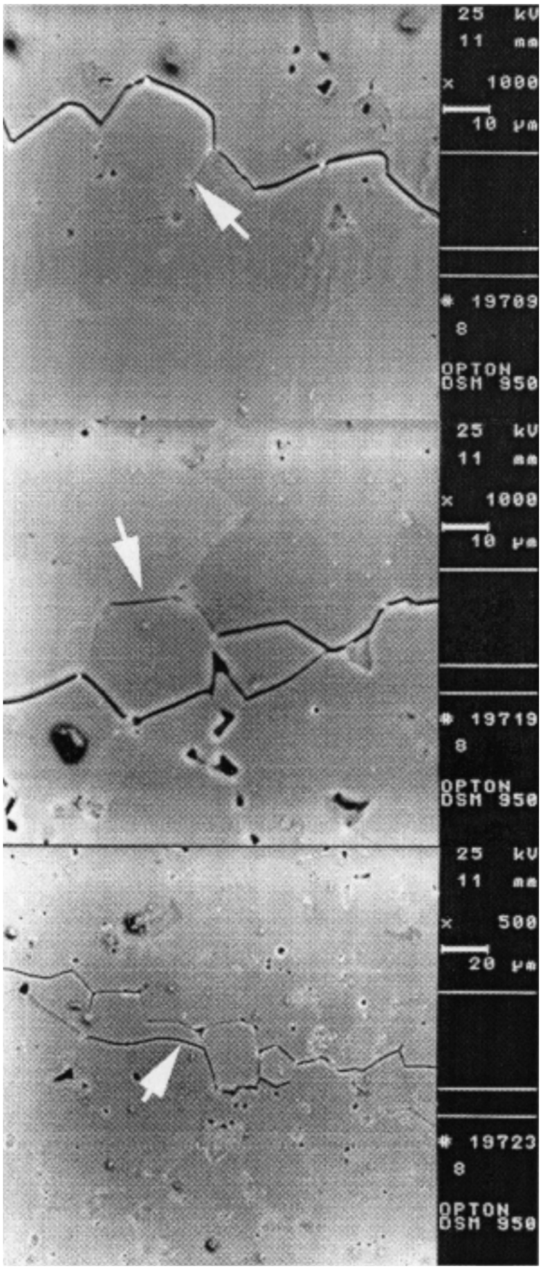

Fig. 1. Single and a group of Al_2O_3 grains acting as bridging grains, observed in alumina ceramics (bridging grains are shown by white arrows).

the amount of bridging grains on Al_2O_3 grain size was also found. From Table 1 it can be seen that the part (%) of a whole path length of the crack propagating through an alumina ceramic and which surrounds a bridging grain, increases with increasing size of the alumina grains.

As was stated by Bennison et al.,[1] the bridging grains are expected to influence the stress intensity factor, K_I, as the crack grows. The data of $K_I = f(c)$ obtained in the range of crack length studied were fitted by a liner equation $y = ax + b$ and the slope a was used as a factor describing the R-curve behaviour of ceramics. Values of parameters a and b in a function of grain size are listed in Table 1. The slope parameter a strongly increases from 0.44 to 1.89 as the Al_2O_3 grain size increases from 2.6 to 67.4 μm (Table 1 and Fig. 2).

Decrease of Al_2O_3 grain size affects not only the amount of bridging grains but also the mean value of residual stresses generated by thermal expansion anisotropy of alumina (see Table 1). Although the sum of stresses over the material must be zero, variations in stress from one grain to another cause both line shift and a broadening. In a result it is possible to calculate first the average values of the line shift, $\Delta\nu$, and then the mean values of residual stresses present in the ceramics studied. Table 1 shows that the mean stress measured by piezospectroscopy strongly decreases with Al_2O_3 grain size. It means that a decrease of frictional forces acts during grain pullout of the embedding matrix, resulting in a decrease of R-curve behaviour (see values of slope a in Table 1) of alumina ceramics. The stress seems to be sufficiently large only for coarser grain sizes to have a significant effect on the crack propagation path during fracture. In the case of smaller grain size the average stress is noticeably smaller probably due to diffusional flow relaxing the stresses, which can be related to greater grain boundary diffusivity at the smaller grain.[6]

The large thermal expansion mismatch between SiC particles and the Al_2O_3 matrix was thought to create additional regions of tension. This expectation has been confirmed by piezospectroscopic measurements. As can be seen from Table 2, an increasing content of SiC grains changes the residual stresses in alumina matrix to tensile. This in turn should reduce the effectiveness of grain bridging. The dependency of the parameters a and b on SiC content for various composites is listed in Table 2. The value of a for a pure alumina matrix is 0.851, which means that the toughness increases strongly namely from 3.6 to about 5.8 MPa $m^{1/2}$ as the crack length increases up to 4.0 mm. The increase in toughness becomes less pronounced as the SiC content increases. For a SiC content of 30 wt.% in an alumina matrix, the slope parameter decreases almost to zero. The negative effect of SiC addition on R-curve behaviour can be related to the observed change of the residual stresses in alumina matrix. However this relation is not so obvious. As can be seen from Table 2, the presence of SiC particles in alumina matrix effectively inhibits Al_2O_3 grain growth. In a result Al_2O_3 grain size decreases from 4.67 μm for alumina matrix to 0.83 μm. for composite with 30 wt.% SiC content. Parallel with decreasing Al_2O_3 grain size, a strong decrease of the amount of bridging grains in alumina matrix is observed (see Table 2).

In contrast to SiC, the higher thermal expansion coefficient of ZrO_2 was expected to create additional regions of compression in alumina matrix. It has been confirmed by piezospectroscopic measurements. As can be seen from Table 3, an increasing content of ZrO_2

Table 1
Amount of bridging grains, linear coefficients a and b (from equation $y = ax + b$), and mean values of residual stresses measured in alumina ceramics in a function of Al_2O_3 grain size

Al_2O_3 grain size (μm)	2.6±0.8	18.6±8.9	28.4±13.7	39.7±17.2	67.4±29.4
Part of a crack path length surrounding bridging grains (%)	3.4	10.7	24.8	40.3	64.9
a	0.44±0.19	0.66±0.03	1.02±0.09	1.60±0.03	1.89±0.14
b	3.17±0.45	3.17±0.08	1.94±0.09	0.80±0.28	0.13±0.27
Mean value of residual stress (MPa)	63.1±11.8	78.9±15.8	118.4±27.6	110.5±31.5	126.3±43.4

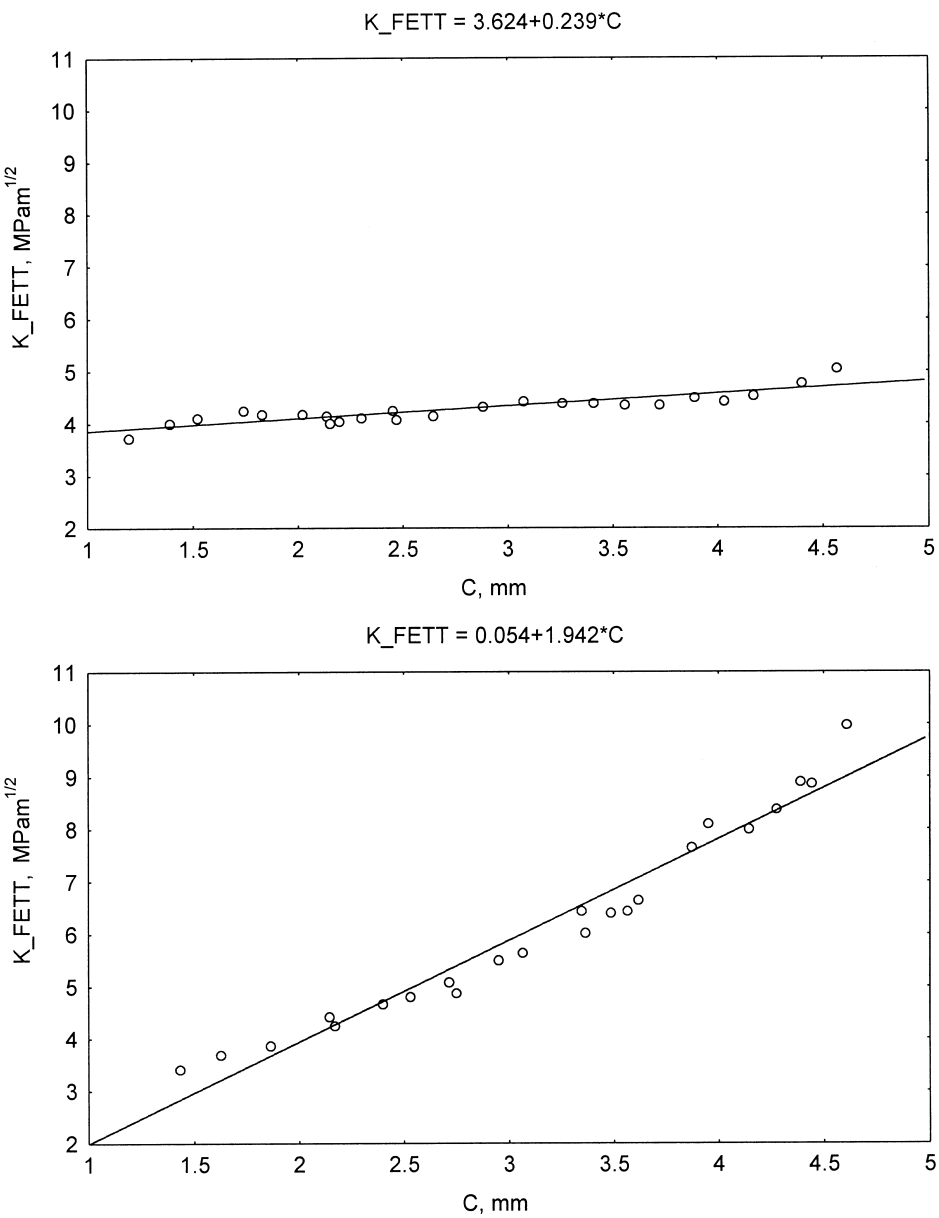

Fig. 2. Dependence of K_I on the crack length c for alumina ceramics with 2.6 μm (top) and 67.4 μm Al_2O_3 grain size (bottom).

grains changes the residual stresses in alumina matrix to compressive. This change should increase the effectiveness of grain bridging. However, controlled crack growth tests showed rapid decrease of R-curve behaviour of composites with increasing zirconia content (Table 3). The slope a equalled 0.665 for alumina matrix decreases to zero for 30 wt.% zirconia content. This intriguing observation can be related to Al_2O_3 grain size change for composites containing zirconia grains. As it was found (Table 3) zirconia strongly inhibits alumina grain

Table 2
Residual stresses, σ (MPa), measured in Al_2O_3 grains, linear coefficients a and b (equation of $y = ax + b$), mean Al_2O_3 grain size (μm) and amount of bridging grains measured in alumina matrix as a function of SiC content

SiC content (wt.%)	0	10	20	30
Residual stress (MPa)[a]	−188.3±35.2	+12.5±2.1	+77.7±14.8	+168.6±30.8
a	0.851±0,074	0.378±0.025	0.224±0.007	0.061±0.024
b	2.64±0,09	3.88±0.08	4.02±0.02	4.44±0.11
Mean Al_2O_3 grain size, μm	4.67±3.91	1.62±0.78	0.96±0.48	0.83±0.27
Part of a crack path length surrounding bridging grains (%)	5.1	0.5	0.1	0.0

[a] Refers to a residual compressive stress (for crystallographic plane *ab* of α-Al_2O_3), + refers to a residual tensile stress.

Table 3
Residual stresses, σ (MPa), measured in Al_2O_3 grains, linear coefficients a and b (equation of $y = ax + b$), mean Al_2O_3 grain size (μm) and amount of bridging grains measured in alumina matrix as a function of ZrO_2 content

ZrO_2 content (wt%)	0	10	20	30
Residual stress (MPa)	78.9±15.1	−81.68±16.4	−148.80±26.3	−194.99±34.6
a	0.665±0,025	0.067±0.011	0.031±0.014	−0.066±0.024
b	3.17±0,08	3.01±0.12	2.93±0.02	2.76±0.11
Mean Al_2O_3 grain size (μm)	18.60±8.90	2.07±1.06	2.22±0.88	1.80±0.46
Part of a crack path length surrounding bridging grains (%)	10.7	0.4	0.05	0.0

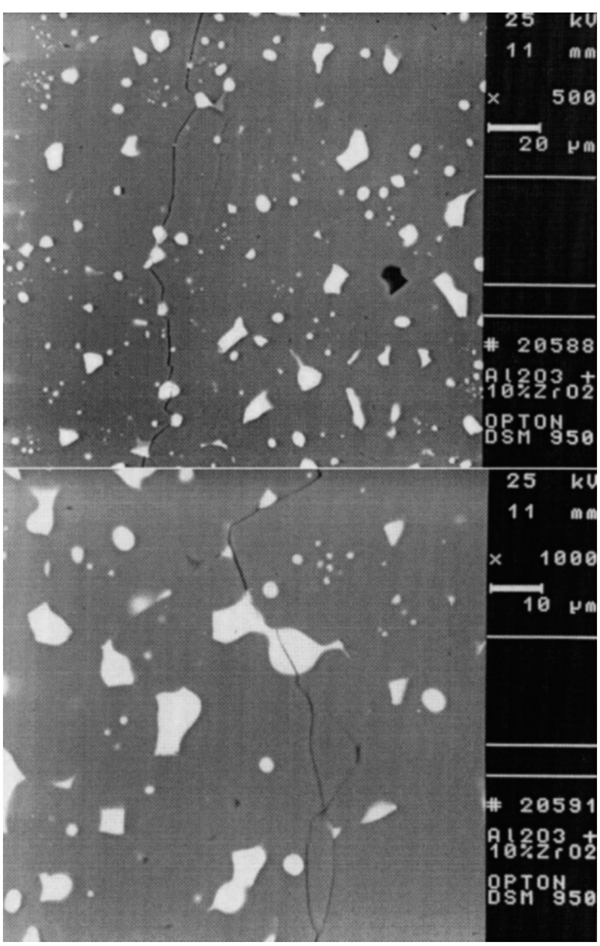

Fig. 3. An example of bridging grains observed in annealed Al_2O_3–ZrO_2 composite.

growth. As a result Al_2O_3 grain size decreases from 18.6 μm for alumina matrix to 1.80 μm for composites with 30 wt.% ZrO_2 content. Similarly to Al_2O_3–SiC composites, rapid decrease of the amount of bridging grains in alumina matrix for Al_2O_3–ZrO_2 composites is also observed. This way, although additional fields of compressive stress are present in alumina matrix it does not prevent an observed decrease of the R-curve behaviour of composites containing ZrO_2. It also means that for fine microstructures, Al_2O_3 grain size is a stronger parameter than internal stresses introduced by a second phase in controlling R-curve properties of ceramics.

To explain the role of residual stresses in phenomenon studied, Al_2O_3–ZrO_2 composite samples with the same Al_2O_3 grain size as for alumina matrix were prepared by annealing initial samples and the tests of controlled crack growth were performed. As is shown in Table 4, the slope parameter a for a composite containing 10 wt.% ZrO_2 is really higher than the same obtained for

Table 4
Comparison of linear coefficients a and b (equation of $y = ax + b$) and amount of bridging grains in alumina ceramics and Al_2O_3–ZrO_2 composite having the same average Al_2O_3 grain size

Type of ceramics	Alumina	Al_2O_3–ZrO_2 composite
Zirconia content (wt%)	0	10
Mean Al_2O_3 grain size (μm)	18.60±8.90	18.96±9.56
a	0.665±0,025	1.087±0.174
b	3.17±0,08	2.19±0.37
Part of a crack path length surrounding bridging grains (%)	10.7	11.02

alumina ceramics. However, the amount of bridging grains is equalled for both ceramics. It means that the observed increase in slope *a* is a result of residual stresses in alumina matrix introduced by zirconia. An example of bridging grains found in a path of the crack propagating through the annealed composite sample is shown in Fig. 3. The results obtained point out that for coarser microstructures where an increased amount of bridging grains is observed, internal stresses as well as Al_2O_3 grain size are variables which control the level of toughening achievable via the grain bridging mechanism.

4. Conclusions

The aim of this work was to determine the effect of grain size and residual stresses on R-curve properties of alumina ceramics and alumina based composites. To realise the first part of the work, alumina samples with grains in the size range 2.6–67 μm were prepared. Residual stresses, generated by thermal expansion anisotropy of Al_2O_3 and dependent on grain size, were found in ceramics. Tests of controlled crack growth for all samples were performed. Data of $K_I = f(c)$ obtained in the range of crack length studied were approximated by a linear equation of $y = ax + b$. As a result the slope parameter *a* was used as a factor describing the R-curve behaviour of ceramics. Decreasing slope *a* with a decreasing Al_2O_3 grain size was found. Observed changes were related to the amount of bridging grains on the path of the propagating crack and residual stresses present in ceramics.

To facilitate the second part, SiC particles, having a lower thermal expansion coefficient, α, or ZrO_2 particles with higher α, were added. A decreasing effect of SiC and ZrO_2 particles content on toughness-crack length dependence was found. The observed changes were related to measurement of alumina grain size, amount of bridging grains and residual stresses. Both additives strongly decreased the matrix grain size. The amount of bridging grains in both cases dropped almost to zero. However, increasing content of SiC grains changed the residual stresses in alumina matrix to tensile but ZrO_2 to compressive. Analysis of the results obtained pointed to Al_2O_3 grain size as a stronger parameter than residual stresses in decreasing R-curve behaviour of alumina based composites with fine microstructures.

To explain the role of residual stresses in the phenomenon studied Al_2O_3–ZrO_2 composite samples with the same Al_2O_3 grain size as for the alumina matrix were prepared by annealing initial samples. It was found that the slope parameter *a* for composite containing 10 wt.% ZrO_2 is really higher than the same obtained for alumina ceramics. The same amount of bridging grains found in both ceramics means that the observed increase in slope *a* is a result of residual stresses in alumina matrix introduced by zirconia.

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