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Mechanical Properties of Polytypoidally Joined Si_3N_4-Al_2O_3

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ABSTRACT

Crack-free joining of alumina and silicon nitride has been achieved by a unique approach introducing sialon polytypoids as a Functionally Graded Materials (FGM) bonding layer. The polytypoid compositions are identified in the phase diagram of the Si_3N_4-Al_2O_3system. The thermal stresses of this FGM junction were analyzed using a finite element analysis program (FEAP) taking into account both coefficient of thermal expansion (CTE) and modulus variations. From this analysis, the result showed a dramatic decrease in radial, axial and hoop stresses as the FGM changes from three layers to 20 graded layers. Scaling was considered, showing that the graded transition layer should constitute about 75% or more of the total sample thickness to reach a minimal residual stress. Oriented Vickers indentation testing was used to qualitatively characterize the strengths of the joint and the various interfaces. The indentation cracks were minimally or not deflected at the sialon layers, implying strong interfaces. Finally, flexural testing was conducted at room temperature and at high temperature. The average strength at room temperature was found to be 581 MPa and the average strength at high temperature (1200 ^{o}C) was found to be 262 MPa. Scanning electron microscope observation of fracture surfaces at a different loading rates indicated that the strength loss at higher temperatures was consistent with a softening of glassy materials present at grain junctions.

Key words: Functionally Graded Material (FGM), Sialon Polytypoid, thermal stress and three- point bend test

1. INTRODUCTION

Joining of high temperature structural ceramics to other materials has often been a significant technological challenge. A commonly used joining technique for similar ceramics involves the introduction of some bonding interlayer. However, the simple bond layer approach is ineffective for dissimilar materials when the thermal expansion coefficients (CTEs) of the materials to be joined are substantially different, since then large stresses arise that cause failure. The possibility of using a graded junction, rather than an abrupt bond layer allows for potentially effective joining of ceramics with widely differing CTEs. In such a graded bond layer, *i.e.* an FGM bond, there is a continuous change in composition from one side to the other, with an accompanying compatible gradient of thermal expansion properties. This concept has been used successfully in sialon polytypoidal functional gradient joining of dissimilar ceramics, Si_3N_4 and Al_2O_3, described in ref [1].

In this paper, we report on the various mechanical properties of the joined ceramics that have been investigated by (a) oriented Vickers indentation test, (b) computation of residual stresses by a finite element method, and (c) strength testing at room and at high (1200 ^{0}C) temperatures.

Many of the present-day advanced materials are composites incorporating various types of dissimilar reinforcing elements such as fibers, whiskers, particles, *etc.*, embedded in a matrix material, introducing a high density of interfaces. To study the mechanisms of how cracks behave along such interfaces, oriented indentation tests have been used [2]. He and Hutchinson analyzed [3] considered the energies for deflection and penetration of a crack through the interfaces, and showed that the toughness of the interface may be determined from the critical angle at which the transition from crack penetration to deflection occurs, with a lower angle of transition implying a tougher interface [4].

As in many joining and composites problems, the effect of residual stress, arising either from processing or from in-service temperature variations, takes on an important role. Since one critical design goal and motivation in FGM research is the minimization of such thermal stresses, several studies have focused on the theoretical and experimental assessment of these stresses in FGMs. A majority of this analytical work has been for FGM films or other simple structures, for which geometrical assumptions allow for much simplified 1-D linear elastic calculations [5]. For a more general 2-D or 3-D problem, numerical methods such as a finite element analysis (FEA) are required. In this work, residual stresses were analyzed in the axisymmetric mode, using the computer program FEAP [6].

Silicon nitride is a candidate ceramic for structural use because of its high strength and toughness. Sialon polytypoidal functional gradients have been used to join the dissimilar ceramics, Si_3N_4 and Al_2O_3 since sialon polytypoids are physically and chemically compatible with both Si_3N_4 and Al_2O_3. In this work, room temperature and high temperature (1200 ^{o}C) strength is evaluated on this ceramic joint [7].

2. EXPERIMENTAL PROCEDURE

(1) Material Fabrication

The samples were prepared by successively filling a graphite hot-press die with the appropriate layers of powder mixes. The sample was subsequently hot pressed at 50 MPa, at 1700 ^{0}C, for two hours, and furnace-cooled to room temperature at 2^{0}C/min. Bend beams and microscopy samples were prepared from these hot-pressed specimens. Details of the processing and characterization methods are those described by Lee *et a.l*, Ref. 1. After hot pressing, each layer had a thickness of 0.5 mm.

(2) Oriented indentation test

Indentation techniques were used to characterize qualitatively the integrity of the joint. Samples were prepared by cutting the FGMs, and polishing to a surface finish of 1 μm. A Vickers indenter initiated cracks in the vicinity of the interfaces at shallow and high incident angles relative to the joining layer interfaces. The indenter loads ranged from 9 to 5kg, with the lower loads applied on the softer, alumina-rich area side of the joint. Evidence of the possible interaction between the crack and the joining interfaces and qualitative information about the strength of the joint was sought using optical microscopy.

(3)Calculation of residual stresses using the Finite Element Method Program

The thermal stresses of this FGM can be analyzed taking into account both CTE and modulus variation of the multitude of joining layers. The residual stresses were computed with a finite element method: the FEAP program [6]. A mesh of 600-4500 nine-node quadrilateral elements was constructed to analyze the problem in axisymmetric mode. A special finite element was formulated so that the Young's moduli and the CTEs varied in the z-direction within each element. Figure 1 shows the coordinate system, while Table 1 lists the materials properties used to calculate the stress. The material composition is taken to be constant in both the radial and circumferential directions. The comparison in residual stress between abrupt joining, *i.e.* without a gradient, and the 20 layer FGM was evaluated. Additionally, the FEAP assessed the dependence of the stresses on the thickness of the graded region, x, relative to the overall sample thickness, T.

(2) Strength Characterization

Three-point bend tests were conducted at 25 ^{0}C and at 1200 ^{0}C to determine the effect of temperature on the strength of the joint. The sample location and test geometry are shown in Figure 2. The test jig was designed specifically for these bend specimens which were 2 mm x 4 mm x 10 mm; the test span form the center to the outer load point was 4 mm; the load displacement rate was 0.06 mm/min. Some tests were also done at a load displacement rate of 6mm/min. Tests at high temperature were carried out in Argon, at 1 atm. The fracture surfaces were examined in the scanning electron microscope (SEM).

3. RESULTS AND DISCUSSION

(1) Joint microstructure

A transmission electron micrograph of Al_2O_3-rich area of the FGM joint of a sample with 20 layers is shown in Figure 3. This micrograph shows that no undesirable reaction took place between the Si_3N_4, the Al_2O_3, or the polytypoids as predicted in the phase diagram [1]. In this region, no phases other than Al_2O_3 and 15R, were detected so that the rule of mixture could be used to design the gradient. The intergranular materials were found to be yttrium aluminates resulting from the use of Y_2O_3 as a sintering additive. Generally, the shape of Al_2O_3 grains was equiaxial with the size ranging from 3 to 10 μm. 12H and 15R polytypoids showed an elongated grain size, 0.5 to 0.9 μm in width and 3 to 6 μm in length. The grain shape of Si_3N_4 is mostly elongated with a grain size about 0.3 μm in width and 2.5 to 4 μm in length.

(3) Indentation test

Figure 4 shows some indentations in the Si_3N_4-rich part of the joint, at the juncture of the Si_3N_4 layer and the first polytypoid layer with composition 90 wt% Si_3N_4/10 wt % 12H Sialon ($Si_3Al_7O_3N_9$), at a high and a shallow incident angle to the layer interface. The interfaces generally could not be discerned in either the SEM or the optical microscope, so the indentations were made close to sporadic interface pores. The indent cracks ostensibly traversed the interfaces at all angles of incidence and were only minimally deflected, implying strong interfaces within the FGM joint. [8,9]

(4) Residual stresses computations

Figures 5 -7 show the computed stress distribution of a 3 layer *vs.* a 20 layer FGM. In the three-layer sample, a 0.1 mm layer of 12H sialon was assumed to join the 19mm diameter cylindrical slabs of silicon nitride to alumina. The 20-layer FGM was assumed to consist of a sequence of 0.5 mm layers with the layer compositions evenly ranging from 100% silicon nitride to 100% alumina, with the intermediate compositions consisting of 18 graded layers with 12H polytypoid sialon mixed with either alumina or silicon nitride (see Reference 1). The stresses were calculated in MPa/^{0}C, with the processing temperature of 1700 ^{0}C assumed to be the temperature of zero stress. Room temperature stress values (20 ^{0}C) *e.g.* are then obtained by multiplying these stresses by 1680. The results show a dramatic decrease in radial stresses as well as in the axial and hoop stresses, as expected, for the 20 layer FGM compared to the tri-layer (Si_3N_4/ 12H sialon/Al_2O_3). The maximum radial stress in Figure 6 shows that it is comparable to maximum axial stresses. For the axial stress at r=R in the tri-layer joint, the range of stress was found to be from +0.9 x 10^5 Pa/^{o}C (compressive) to -1.2 x 10^5 Pa/^{o}C (tensile) whereas in the 20 layer FGM, the range of stress was found to be from 0 Pa/^{o}C to 1.8 x 10^5 Pa/^{o}C (compressive) near the center of the joint. These analyses can give an estimate of the expected residual stress in actual FGM joints and indicate the approximate number of layers necessary to achieve a crack-free juncture [10,11]. The variation of the stresses, σ_{rr}, with R at z=T/2 for the 20-layer sample is shown in Figure 8. σ_{rr} is zero at R=r, and is maximum at r=0, while σ_{zz} is maximum at r=R.

An important consideration for the potential applications of this sample is the range of geometries in which an FGM approach can be useful. Two situations were examined: the joining of equal thicker slabs of silicon nitride to alumina via a 20-layer graded joint of fixed thickness, with the relative thickness characterized by the ratio x/T (see Fig. 10), and an asymmetrical joining of a thin alumina layer to a thick piece of silicon nitride, Figure 9. The resultant calculations for the axial stress at r=R (the outer diameter) show that the stresses rise dramatically as the FGM joint becomes thin compared to total sample dimensions. The example for the symmetrical case show that the axial stress at r=R and z=T/2 scales approximately linearly with the decreasing x/T ratio between x/T=0.75 and x/T=0. Also this figure compares using a fixed thickness for the graded joint *vs.* using the same fixed thickness for the single sialon interlayer (*i.e.* the tri-layer specimen) for abrupt joining between silicon nitride and alumina. There is an increasingly large difference in the maximum stress between graded layer and abrupt joining as x/T ratio increases, indicating the importance of grading the joint to minimize the stress. As x/T values become smaller, however, the graded approach holds minimal or no practical advantage over single-layer joining. Large stresses also arise in the asymmetrical case as the example in the Figure 9 showed. It can thus be concluded that joining of dissimilar ceramics such as silicon nitride with alumina, can only be expected to be successful for situations where the FGM gradient make up 75% or more of the entire sample thickness, limiting severely the range of applications of the FGM approach to joining.

(4) Strength Characterization

Flexural strengths at room temperature and at 1200 ^{o}C were obtained using a three-point bend test. The fracture for both temperatures occurred within the polytypoid which is approximately in the middle of the sample. The average strength at room temperature was found to be 581 MPa and the average strength at 1200 ^{o}C was found to be 262 MPa (Table 2). 4-5 samples were tested at each temperature. Fracture surfaces of the tested samples at room temperature and at high temperatures were examined by SEM. Figures 11 and 12 show that at both room temperature and at high temperature, the fracture modes are mixed intergranular and transgranular. The fracture surfaces of the materials tested at 1200 ^{o}C show evidence of viscous deformation of intergranular material, Figure 13. This observation agrees with the previous work of Cinibulk et. al. (1990) on Si_3N_4 sintered with $Y_2O_3 + Al_2O_3$ which reported grain-boundaries with poor resistance to softening at 1000 ^{o}C [12,13] and that done by Li et.al [7]. In these liquid-phase sintered ceramics, grain boundaries and triple-junctions often contain an amorphous phase which can soften, resulting in ready grain-boundary sliding, cavitation, and cracking. To confirm the observation of softening of the glassy phases in the FGM joint, a loading rate of 6 mm/min was used at 1200 ^{o}C. The flexural test using 6 mm/min loading rate at 1200 ^{o}C resulted an average strength value of 356 MPa with a standard deviation of 36 MPa compared to 262 MPa at the slow loading rate. This confirms that the strength degradation at high temperature is most likely due to the softening of intergranular glassy phases. While high resolution images of various grain boundaries did not detect grain boundary glassy phases (see ref [1]), significant quantities of glass resides at triple points. A combination of grain boundary sliding with triple point glass softening may thus be proposed as the origin of the loading-rate dependent modulus of rupture of the FGM joint. Further alloying to crystallize these grain boundary phases could be beneficial to improved high temperature performance [12-14].

5. CONCLUSION

Crack free joining of heterogeneous ceramics is demonstrated by the use of sialon polytypoids as Functionally Graded Materials (FGM) as defined by the phase diagram in the system Si_3N_4 – Al_2O_3. Based on the fabrication method and mechanical characterization obtained by indentation testing, residual stress calculations using FEAP, and strength testing, the following conclusions can be made:

(1) The indentation test at high and shallow incident angles, shows that cracks pass through the interface without being deflected, which is evidence that the interface is strong.

(2) FEAP indicates the stress distribution in the FGM sample. The result showed a dramatic decrease in radial, axial and hoop stress as the FGM changes from three layers to 20 graded layers. This analysis explains why a 20 layer FGM was crack-free but a 3-layer FGM was cracked. Scaling computations for FGM samples indicate that at least 75% of the total sample thickness needs to be the graded thickness so as to minimize residual

stresses. Such analyses are especially useful for graded FGM samples where the residual stresses are very difficult to measure experimentally.

(3) The average strength was found to be 581 MPa and 262 MPa at room and high temperature respectively. The fracture for both temperatures occurred in the middle of the sample. This strength loss at high temperature was consistent withg a softening of glassy phases at triple junctions.

6. Acknowledgement

This work was supported by the Director, Office of Basic Energy Sciences, Division of Materials Sciences of the United States Department of Energy. The author would like to thank Dr. Terrance Becker and Dr. Rowland Cannon at LBNL for help in setting up FEAP program to calculate residual stress. Also, the author would like to thank Dr. Jim McNaney for his help with creep furnace to do strength tests.

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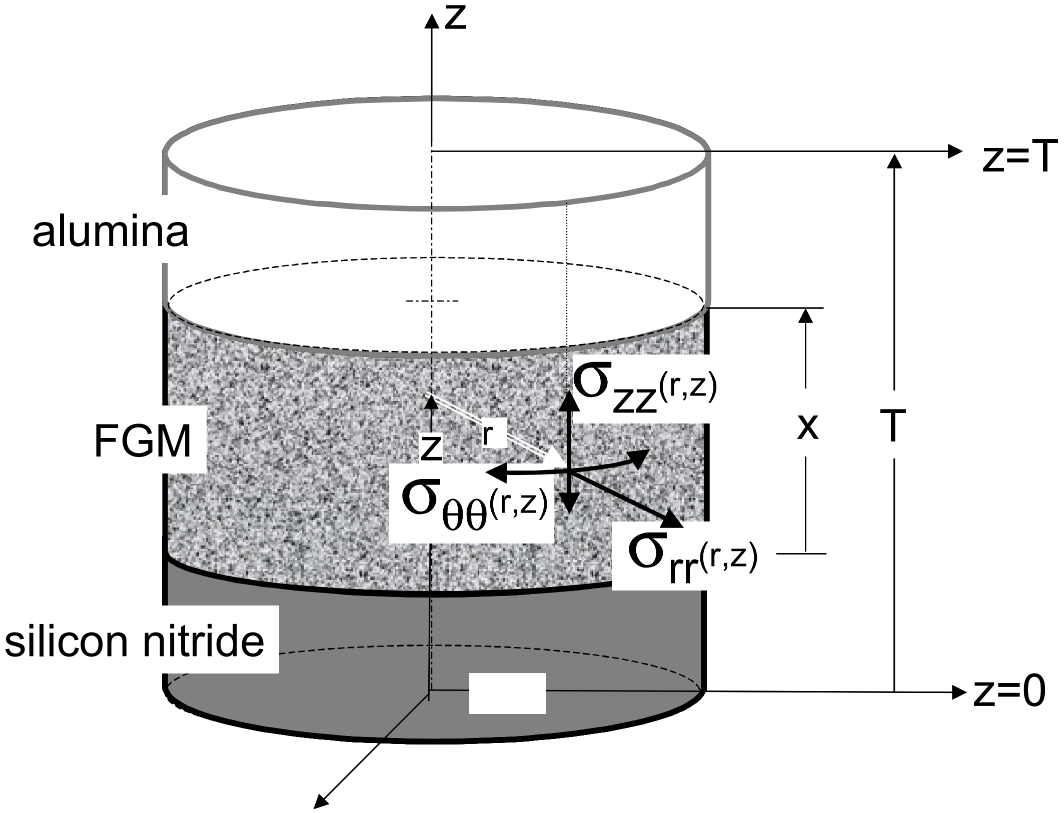


Figure 1. Sample geometry and coordinate systems

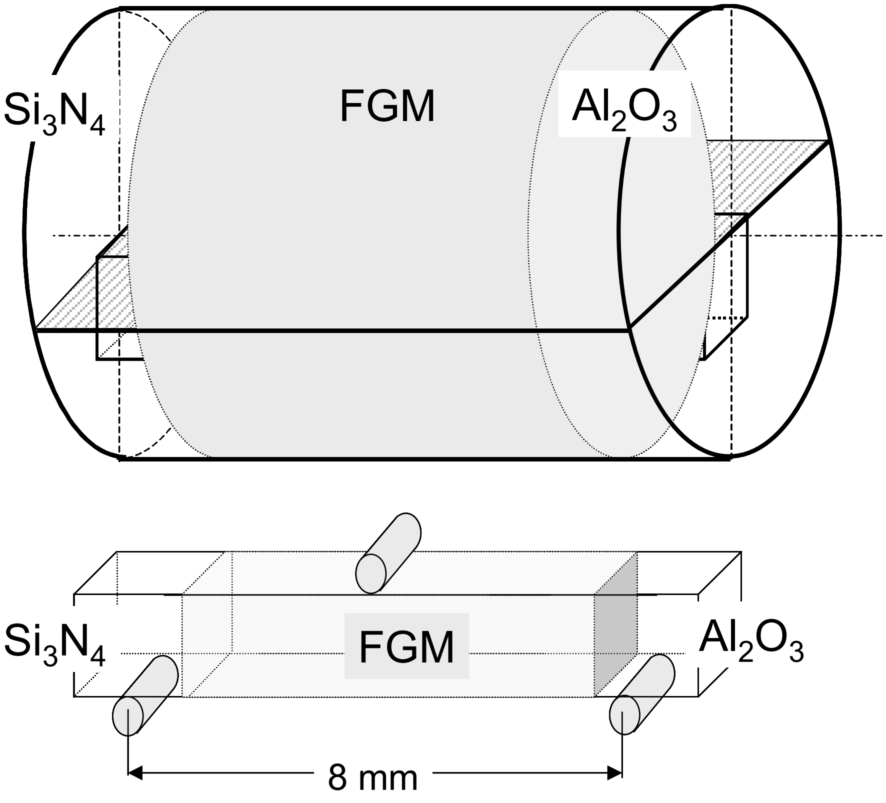


Figure 2. Sample position and test jig geometry.

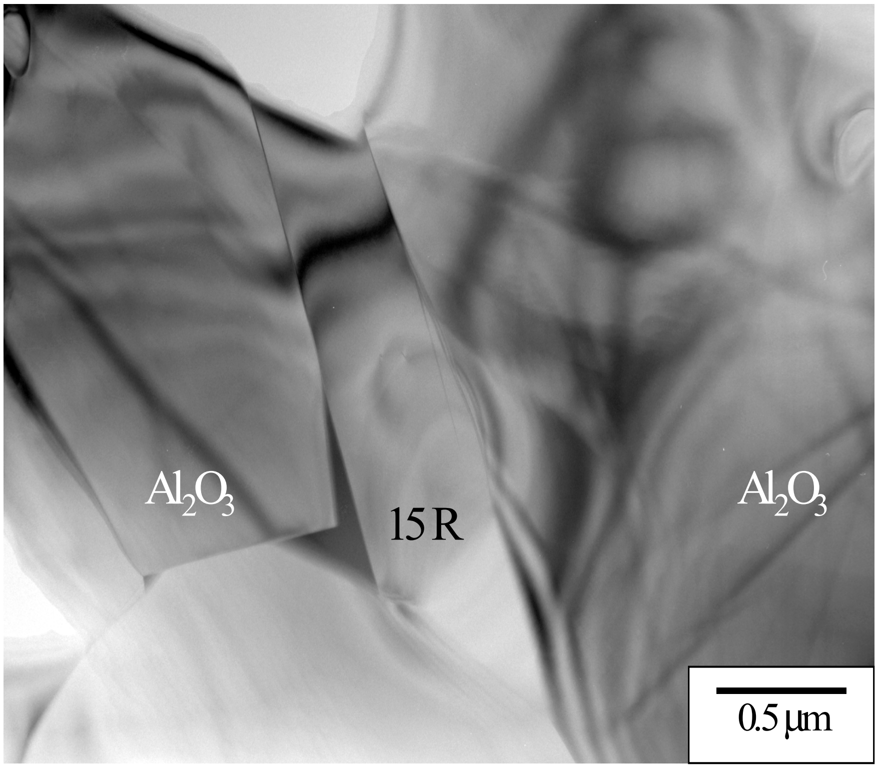


Figure 3. TEM image of the microstructure in the alumina-rich side of the FGM joint. Only alumina and 15R phases were detected, together with some glassy phase at triple junctions.

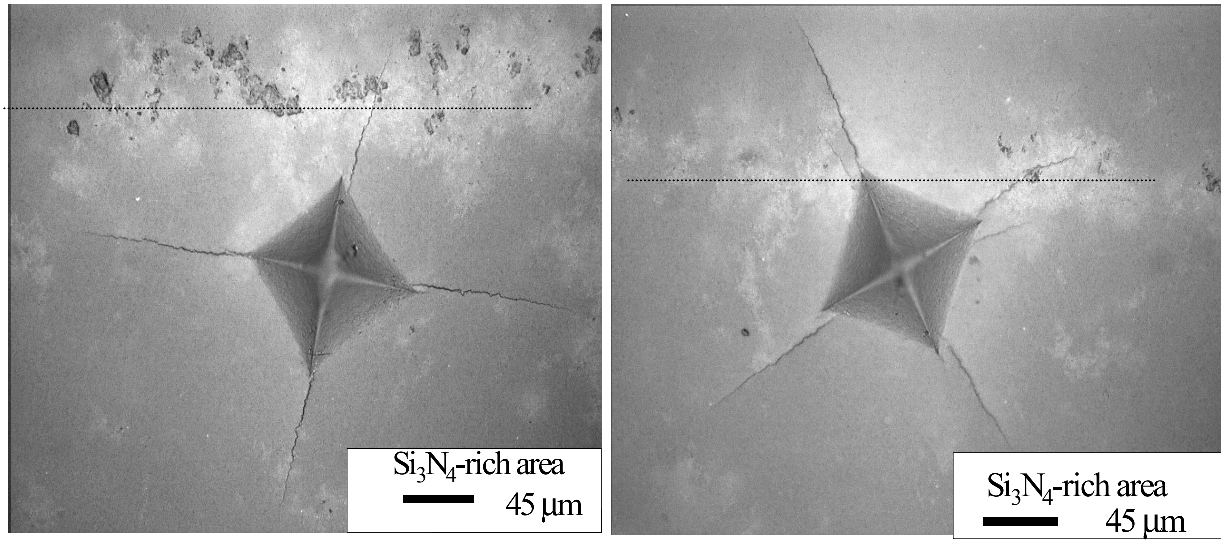


Figure 4. Optical micrographs of the joints showing Vickers indents at high and shallow incident angles in the Si_3N_4-rich area. The cracks pass through the joint without being deflected. The dotted lines indicate the position of the interfaces.

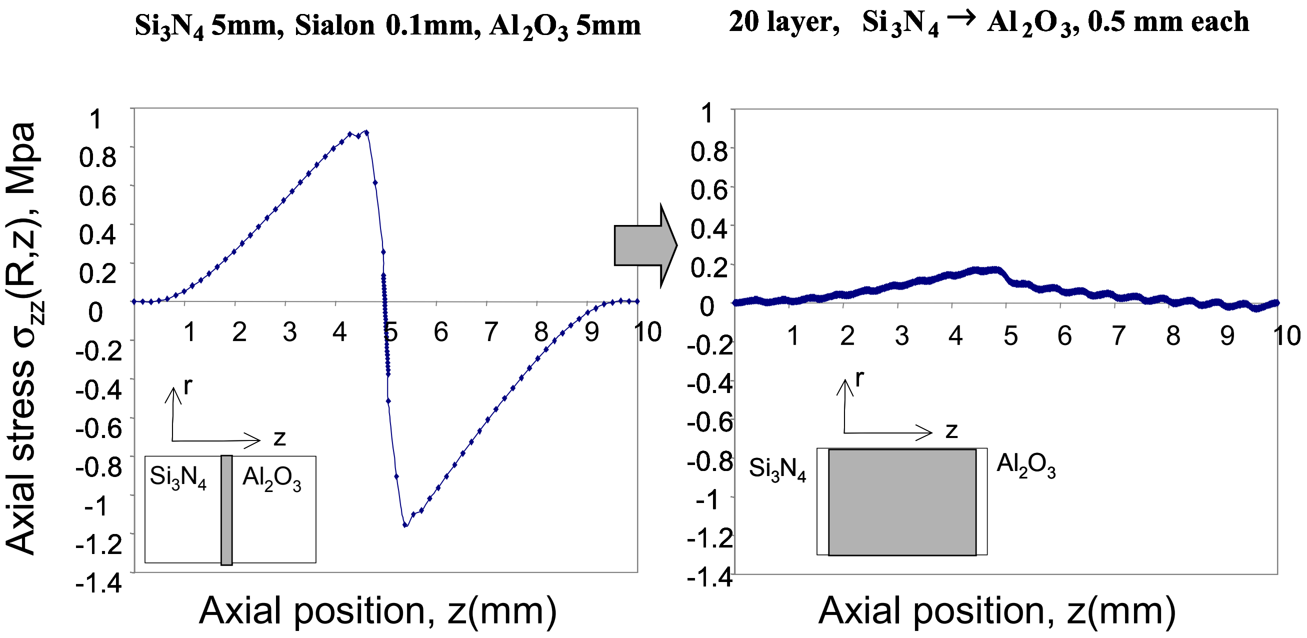
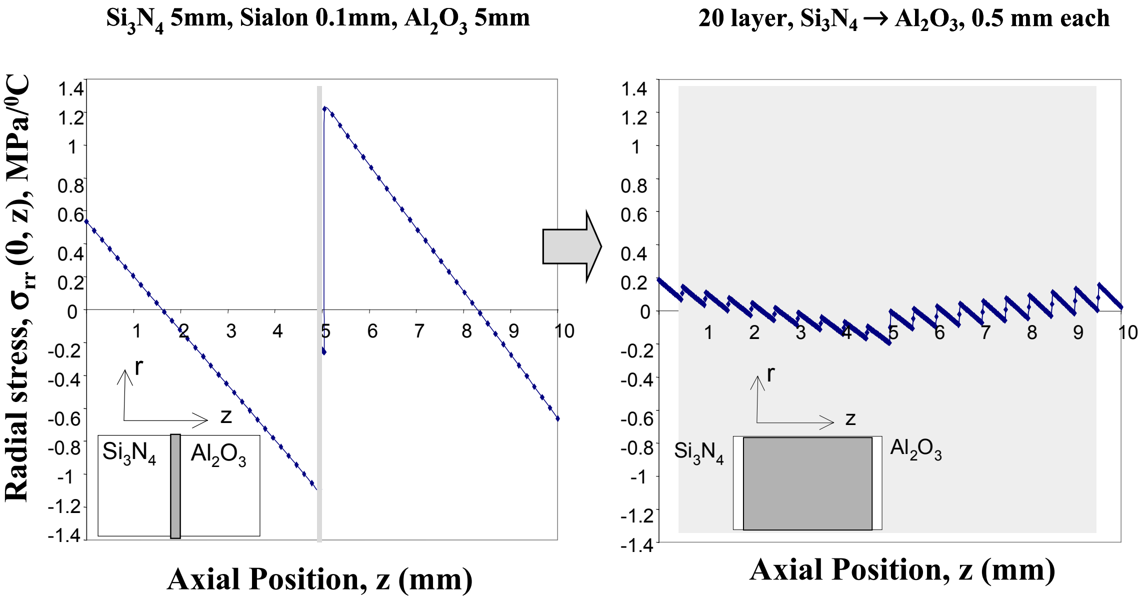
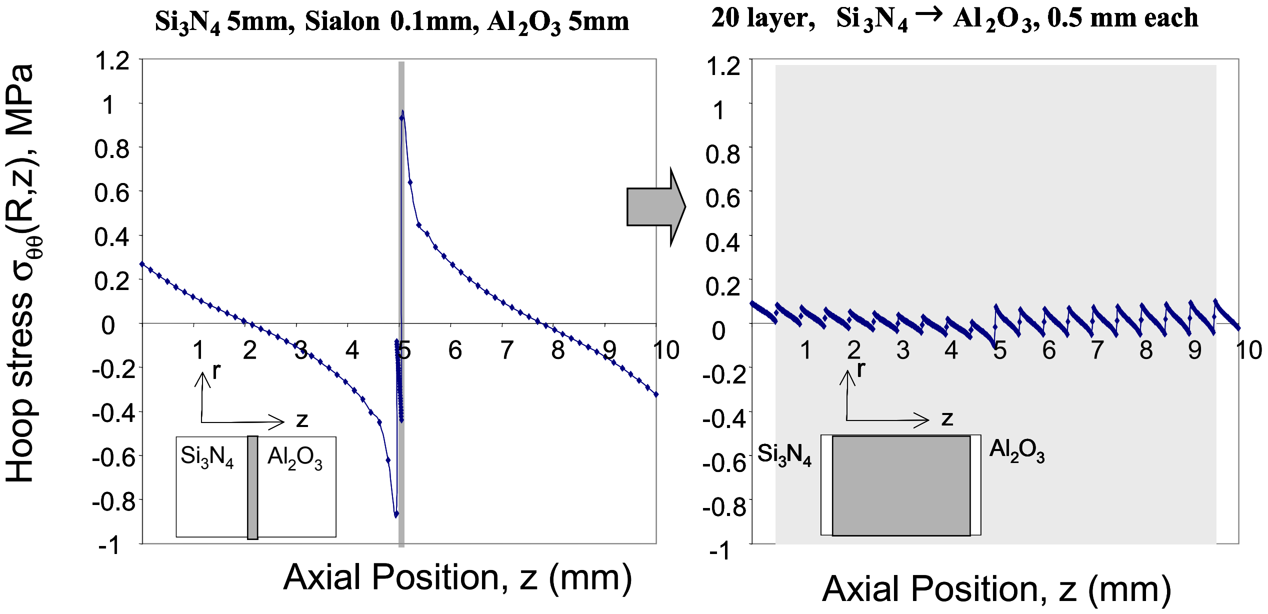


Figure 5. Comparison of the computed axial stress, σ_{zz} at r=R as a function of z, for a 3-layer sample versus a 20 layer FGM cylindrical sample with a 19 mm diameter.


Si_3N_4 5mm, Sialon 0.1mm, Al_2O_3 5mm

20 layer, $Si_3N_4 \rightarrow Al_2O_3$, 0.5 mm each

Figure 6. Computed radial stresses, σ_{rr}, at r=0 as a function of axial position, z, for the tri-layer and the 20 layer FGM samples.



Si_3N_4 5mm, Sialon 0.1mm, Al_2O_3 5mm

20 layer, $Si_3N_4 \rightarrow Al_2O_3$, 0.5 mm each

Figure 7. Computed hoop stresses as a function of axial position , z, at r=R.

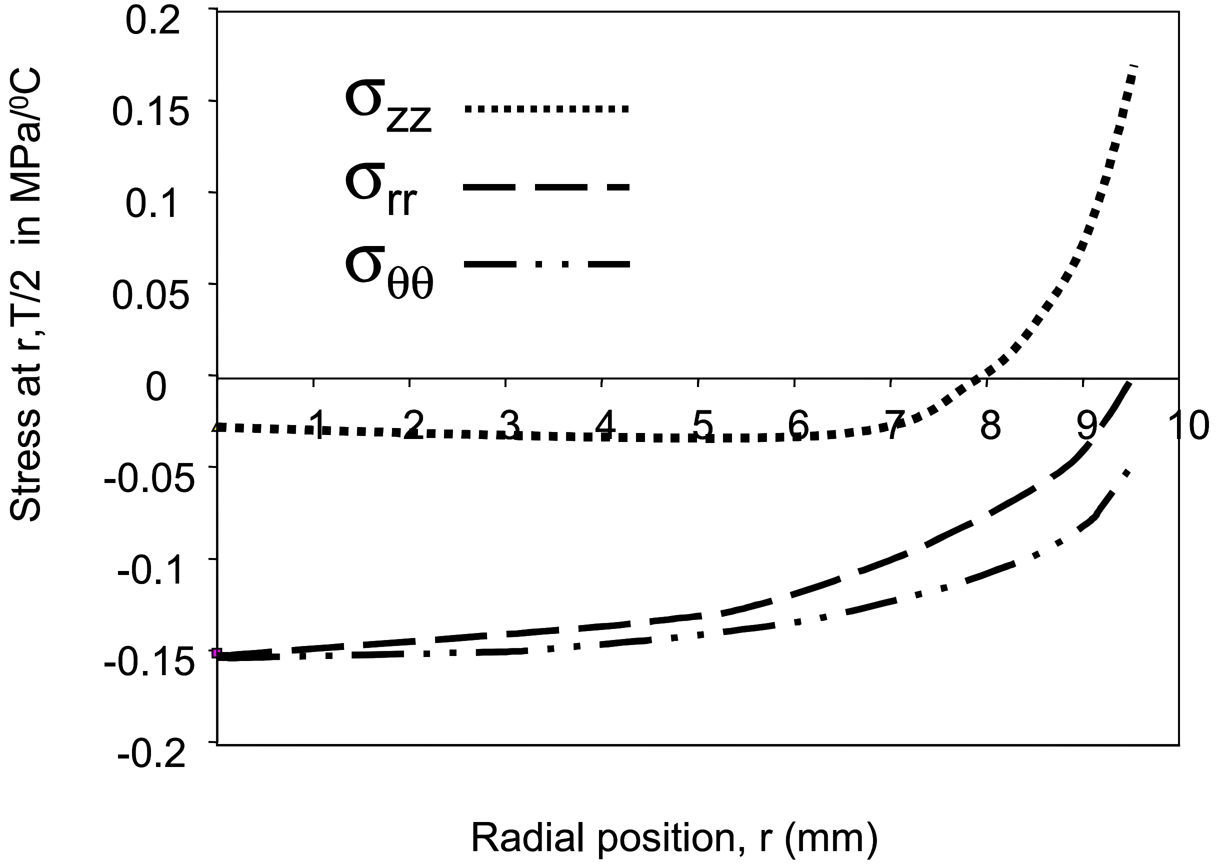


Figure 8. Computed stresses as a function of radial position, r, at z=T/2, for the symmetrical 20-layer FGM sample. The sample diameter is 19 mm; every layer has a thickness of 0.5 mm.

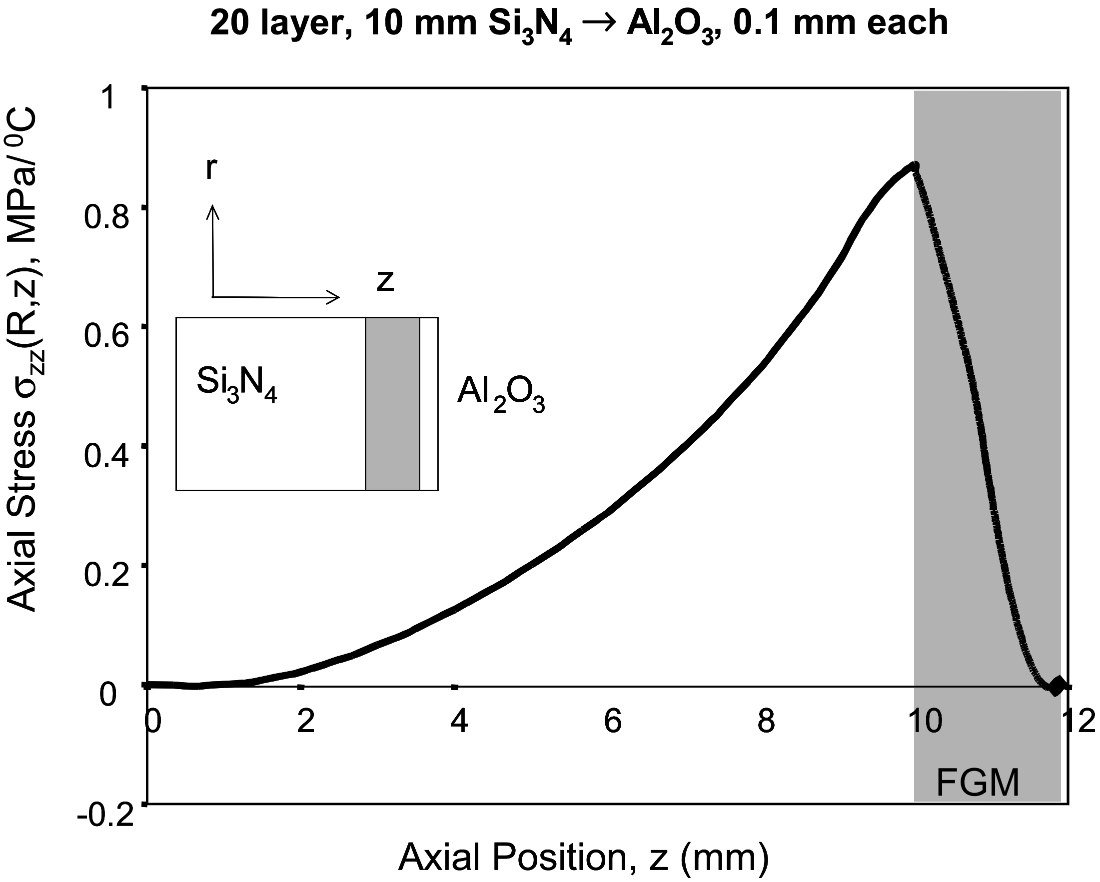


Figure 9. Computed axial stress, σ_{zz} at r=R of asymmetrical FGM sample. The shaded region indicates the position of the FGM.

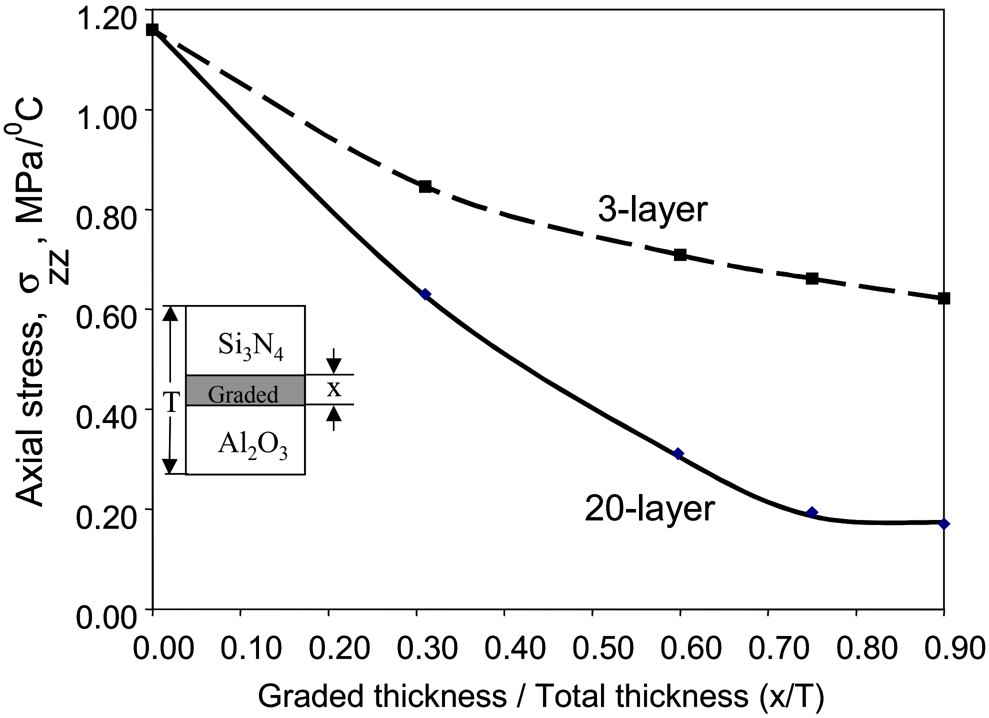


Figure 10. Computed maximum axial stress at r=R, for a fixed joint layer thickness, x, as a function of the total sample thickness T. The silicon nitride and the alumina slabs are assumed to be of equal thickness. As the slab thickness of the alumina and silicon nitride slabs increases, the value of x/T decreases. The actual stress is obtained by multiplying the stress per ^{0}C values by the temperature difference between the stress free temperature (here 1700 ^{0}C) and the evaluation temperature (e.g. 20 ^{0}C). This scaling computation sets the limits of sample geometry for which successful joining may be expected.

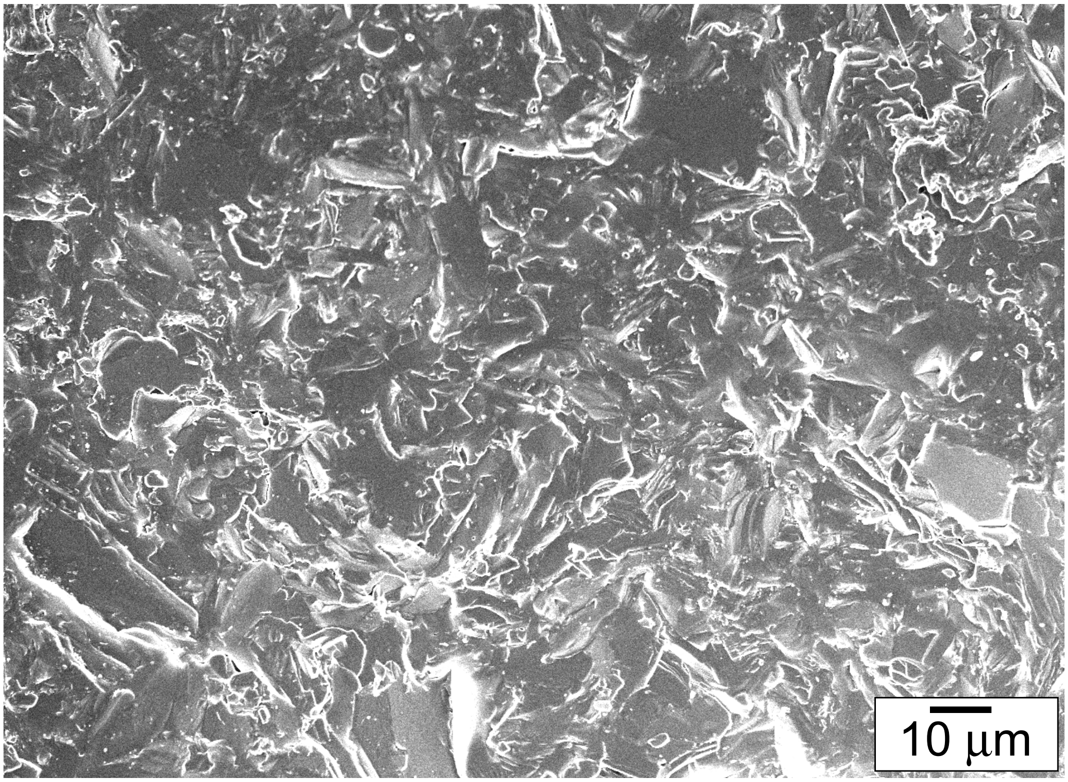


Figure 11. SEM image of room temperature fracture surface at z~ T/2 of a 20-layer FGM sample.

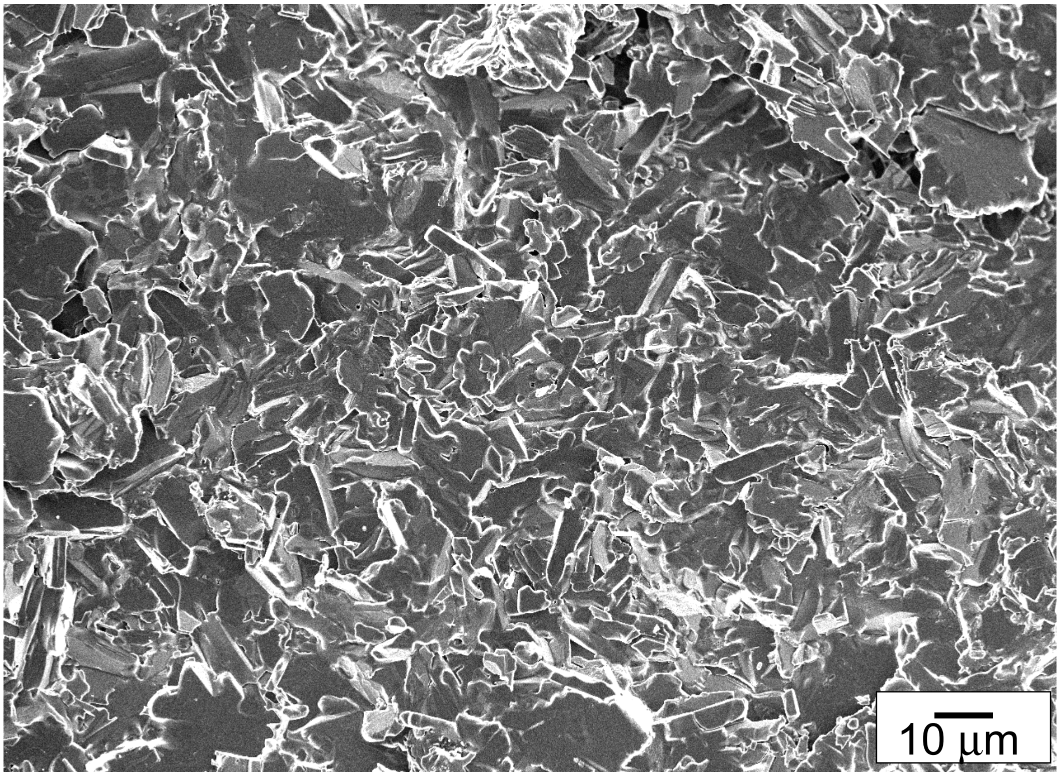


Figure 12. SEM image of 1200 ^{0}C temperature fracture surface at z~ T/2 of a 20-layer FGM sample.

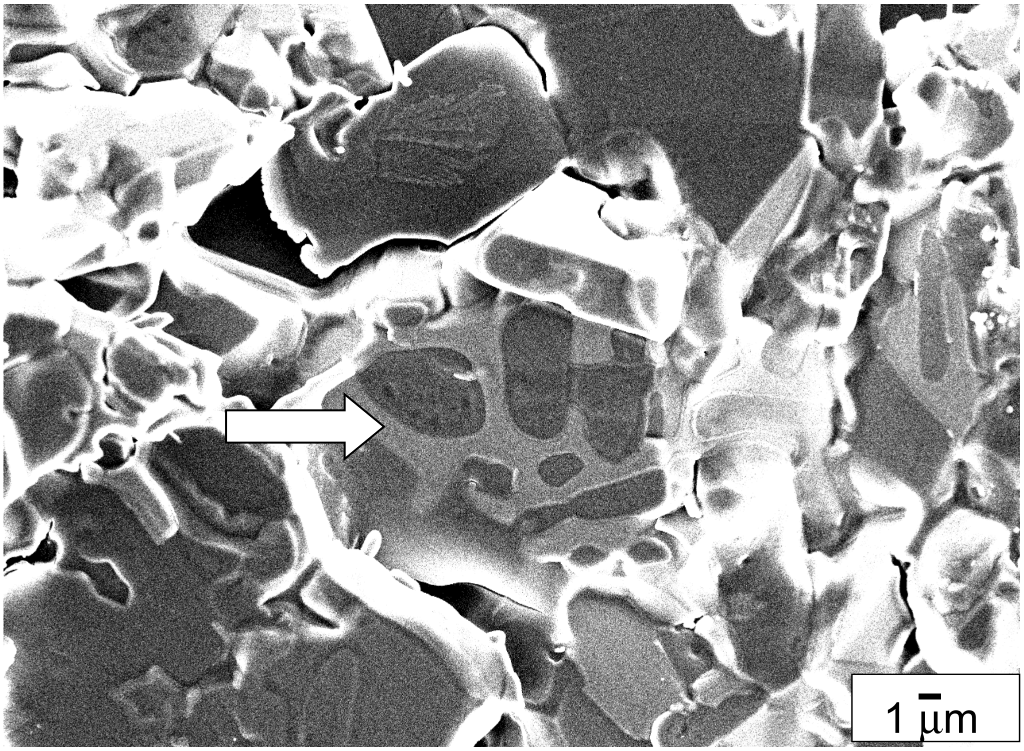


Figure 13. SEM image of 1200 ^{0}C fracture surface with presence of viscously deformed intergranular phase indicated.

Tables.

Table 1. Physical Constants for the materials under study

Properties	Si_3N_4	Polytypoid	Al_2O_3
E (Gpa)	330	290	390
ν (Poisson's ratio)	0.22	0.22	0.22
α (*$10^{-6}/^0C$)	3.6	5.6	8.8

Table 2. Strength test results

Temp/Strength	Strength (MPa)
At Room temperature (25 °C)	581 ± 60
At High temperature (1200 °C) (loading rate 0.06 mm/min)	262 ± 20
At High temperature (1200 °C) (loading rate 6 mm/min)	356 ± 36