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Fabrication of α/β-Sialon Symmetrically Compositional FGM

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Summary:

Sialon as one of the important structural ceramics is being paid more and more attention due to its unique chemistry, crystal structure and microstructure features. α-Sialon has superior hardness, β-Sialon possesses high strength and toughness, and (α+β)-Sialon is combination of α-Sialon and β-Sialon. This paper presents the crystal structure, fabrication and properties of α-Sialon, β-Sialon and (α+β)-Sialon. On the basis of their properties, Symmetrically Compositional Functionally Graded Material (SCFGM) with the advantage of both α-Sialon and β-Sialon, has been fabricated, its thermal stress has been calculated with finite element analysis method, and its microstructure has been investigated.

Keywords:

α-Sialon, β-Sialon, (α+β)-Sialon, symmetric composition functionally graded material (SCFGM)

1. Introduction

There are two well known Sialon phases in the Si_3N_4-rich corner of the R-Si-Al-O-N phase diagrams (R=rare-earth), called α- and β-Sialon, which are isostructural with α and β-Si_3N_4, respectively. The β-Sialons have the composition $Si_{6-z}Al_zO_zN_{8-z}$, with $z \leq 4.2$, along the tie line from Si_3N_4 towards $AlN \bullet Al_2O_3$, while the α-Sialon solid solutions, $R_{m/3}Si_{12-(m+n)}Al_{(m+n)}O_nN_{16-n}$, have two-dimensional extensions in the Si_3N_4- $AlN \bullet Al_2O_3$-$RN \bullet 3AlN$ plane of the corresponding Jänecke prisms[1-3].

α-Sialon ceramics have attracted much attention as structural materials, because they show low linear thermal expansion coefficients(~3.7-4.0×10^{-6}°C) and high hardness(Hv~22GPa), and because they offer the possibility of

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preparing almost glass-free nitrogen ceramics[2, 3]. α-Sialons form equi-axed grains in most case, so they have a high hardness but generally comparatively low fracture toughness. Attempts have been made in the past to reinforced Y-doped α-Sialon ceramics with additions of other refractory compounds like SiC whiskers or $MoSi_2$ particulates. An improved toughness was only partly achieved in the case of SiC whiskers because of too strong bonding between the α-Sialon matrix and the SiC whiskers. The results obtained when using $MoSi_2$ particulates, on the other hand, that the toughness could be increased by more than 50%. However, this was achieved with a concomitant expected loss in hardness; dense $MoSi_2$ ceramics have a hardness of only around Hv=10 Gpa[4, 5]. In order to retain the excellent hardness of the α-Sialon ceramics, other means of increasing the toughness have to be searched for.

Many structural components encounter service conditions and, hence, required materials performance, which vary with location within the component. Functionally graded materials (FGMs) feature gradual transitions in microstructure and composition which are engineered so as to meet functional performance requirements that vary with location within a single component and to optimize the overall performance of the component[6].

As another important member of Sialon race, β-Sialon grains normally grow with elongated shapes together with liquid phase and/or α-Sialon. It is therefore generally accepted that the β-Sialons show an anisotropic grain growth habit while the crack deflection by prismatic β-Sialon grains yield tortuous crack paths which improve the fracture toughness by a energy dissipation mechanism[1-3].

On the basis of the new material concept of FGM, the sintering characters were investigated, and the symmetrically compositional functionally graded material (SCFGM) of α/β-Sialon was fabricated.

2. Experimental

Commercial powder of Si_3N_4, Al_2O_3, Y_2O_3, and AlN are used as raw materials. The characteristics of these powders are listed in Table 1.

Table 1. Characteristics of raw materials

Powder	Particle size (μ)	Purity(%)
Si_3N_4	0.05-1	>97
Al_2O_3	< 1	>99
Y_2O_3	0.1-0.5	>99
AlN	0.5-1	>95

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The amounts of Si_3N_4 crystalline phases were semiquantitative estimated through the X-ray spectrum of Si_3N_4 powder. The weight content of α- Si_3N_4 is 80%, the content of β- Si_3N_4 is 20%. The powders were prepared according to the composition of Sialon, the starting material mixtures were milled in water-free propanol for 6 hours in a plastic jar, and then dried in air.

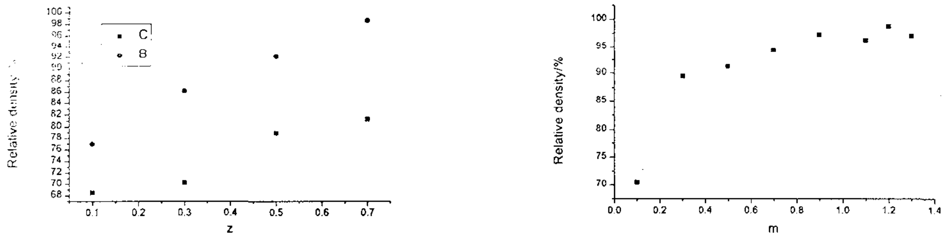
For the Sialon ceramics, pellets of dried powder were compacted in BN-coated graphite dies; for α/β-Sialon FGMs, the starting material mixtures were stacked into BN-coated graphite dies according to the designs of graded layered compositions, and compacted in the die. Then, the green compacts were sintered at a condition of 1780℃, 25MPa and holding for 1 hour in argon. The densities of Sialon ceramics was measured according to Archimedes' method. Scan360 scanning electronic microscopy were used to observe the microstructures of α/β-Sialon FGMs. Thermal stress distribution within FGM was calculated by finite element analysis.

3. Results and discussion

3.1 Sintering of Sialon Ceramics

In order to fabricate the FGMs with excellent properties, the gradient of composition must be chemical compatible and physical matched. Sun[7] has studied the compatible area among the α-sialon phase, M′ phase[$Me_2Si_{3-x}Al_xO_{3+x}N_{4-x}$] and β-Sialon phase. She indicated that the α-sialon phase and M′ phase are compatible, the compatible area of M′ phase and β-Sialon[$Si_{6-z}Al_zO_zN_{8-z}$] phase is $0 \leqslant z \leqslant 0.8$. Slasor[8] has studied the boundary of α-sialon phase and β-Sialon phase. He indicated that the boundary located m=0 and n=0~1.7[$Y_{m/3}Si_{12-(m+n)}Al_{m+n}O_nN_{16-n}$]. In this work, z=0.1, 0.3, 0.5 and 0.7 were selected for the research on the sintering of β-Sialon ceramics. As to the sintering of α-sialon and α-β-Sialon, the samples were prepared along the join line of Si_3N_4-R_2O_3:9AlN in the Si_3N_4-AlN•Al_2O_3-RN•3AlN plane of the corresponding Jänecke prisms, overall compositions with m=0.1, 0.3, 0.5, 0.7, 0.9, 1.1, 1.2 and 1.3[$Y_{m/3}Si_{12-(m+n)}Al_{m+n}O_nN_{16-n}$, n=m/2] in this work. The relative density of Sialon ceramics is shown as Fig.1. Fig.1(a) shows that the relative density of β-Sialon ceramics is raised from 68% to 81% as the z value increases. But the β-Sialon ceramics can't be densified because of too low content of liquid phase in the sintering process. The relative density of β-Sialon ceramics is improved while 2% Y_2O_3 was used as additive, reaching 92% and 98% respectively when z=0.5 and 0.7. So the β-Sialon ceramics can be densified as the increase of liquid phase content in sintering process.

Fig. 1(b) shows that the relative density of α-Sialon and α-β-Sialon ceramics is raised while the m value increased, the α-Sialon and α-β-Sialon ceramics are densified when the m value is higher than 0.5. The relative density is low when the m value is lower than 0.5 because the content of liquid-phase in sintering process is too low. It can be concluded that the relative density is related to the contents of Al_2O_3, AlN and Y_2O_3, which increases as the contents of Al_2O_3, AlN and Y_2O_3 increase.



(a) β-sialon(C—β-sialon, B—Y_2O_3-β-sialon) (b) α-sialon 和α-β-sialon

Fig 1 Relative density of sialon ceramics

3. 2 The phase composition of Sialon Ceramics

Analyses of the crystalline phase of the prepared samples were based on their X-ray diffraction records with Cu $K\alpha_1$ radiation. In the semi-quantitative estimation of the amounts of crystalline phases, the integrated intensities of the following reflection were used: (1) (210) of the α-Sialon, (2) (210) of β-Sialon. The relative fraction of each phase was then calculated from the expression

$$W_\alpha=I_\alpha/I_\alpha+I_\beta$$

$$W_\beta=I_\beta/\ I_\alpha+I_\beta$$

Where W_α and W_β are the fraction of α-Sialon and β-Sialon phase respectively, I_α and I_β represent for the intensity of the reflection on the (210) of the α-Sialon and β- Sialon respectively.

The phase composition of Sialon Ceramics is shown in Table 2. Table 2 shows that the phase composition of Sialon ceramics with compositions z=0.7 and m=0.1 doesn't fit to the Jänecke prisms. It can be concluded that Si_3N_4 powder not only doesn't transform into Sialon ceramics, but also can't transform from α-Si_3N_4 and β-Si_3N_4 completely while the content of liquid phase is too low in sintering process. When the sample was prepared with composition of z=0.7

and 2% Y_2O_3 additive, the single-phase β-Sialon ceramics was obtained. The contents of α-Sialon phase increase as the m value rises. No matter how to raise the m value, the single-phase α-Sialon ceramics can't be obtained with this process.

Table 2 Phase composition of sialon ceramics

Composition	Z=0.7 wt/%	Z=0.7 (Y_2O_3) wt/%	M=0.1 wt/%	M=0.3 wt/%	M=0.5 wt/%	M=0.7 wt/%	M=0.9 wt/%	M=1.2 wt/%
α-Si_3N_4 or α-sialon	23	0	18	13	22	30	43	64
β-Si_3N_4 or β-Sialon	77	100	82	87	78	70	57	36

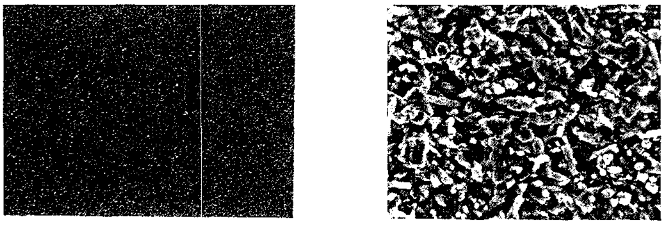
3. 3 Fabrication of α/β-Sialon SCFGM

The goal of Fabrication of α/β-Sialon symmetrically compositional functionally graded material (SCFGM) is to obtain ceramic wear-resistant material with both high hardness and toughness. So the composition of surface layer should be Sialon ceramic with high content of α-Sialon phase, the composition of the central layer is β-Sialon, the content of α-Sialon phase in the graded layers increase from center to surface. The dimension of α/β-Sialon SCFGM is ϕ40mm and 6mm thick. The distribution of composition of α/β-Sialon SCFGM from surface to center is shown as Table 3.

Table 3 Compositions of α/β-Sialon SCFGM

Thickness	1mm	0.5mm	0.5mm	0.5mm	1mm
Composition	M=1.2	M=0.9	M=0.7	M=0.5	β -Sialon
Contents of α phase	64%	43%	30%	22%	0

The microstructure of α/β-Sialon SCFGM is shown in fig.2. Fig. 2(a) shows that the sample is densified completely without obvious interface of graded layers, and no macro-cracks and flaws were found in the sample. Fig.2(b) shows that the amount of elongated grains gradually increases while the amount of equi-axied grains decrease. The distribution of composition of α/β-Sialon SCFGM is no long stepwise as that the composition in distribution of green compact, which is the result of diffusion in the liquid-phase sintering process. The joint between the elongated grains and the equi-axied grains is excellent, the equi-axied grains inlaid among the elongated grains, no micro-cracks and flaws were found in the joints and boundaries.



(a)Scanning of FGM insect (200×) **(b)Micrograph of some areas in FGM (2000×)**

Fig.2 Micrograph of α/β Sialon SCFGM

3. 4 The residual stress of α/β-Sialon SCFGM

Symmetrically compositional functionally graded material is one of the heterogeneous materials, which has symmetrically graded composition distribution and structures. The difference between SCFGM and thermally relaxed FGM is that the latter is designed to reduce residual thermal stress that occurred at the interface between FGM layers, while SCFGM is designed to add residual thermal stress to the SCFGM surface through symmetrical structures. The ideal stress distribution is compressive stress in the surface layer, tensile stress in central layer. The mechanical properties of the two symmetrical surface layers will be improved because of the adding of the residual compressive stress while the central layer with high toughness can withstand the residual tensile stress[9]. But the α/β-Sialon SCFGM is different from the ideal SCFGM, the linear thermal expansion coefficient (CTE) of α-Sialon is higher than that of β-Sialon, so the stress distribution is compressive stress in the central layer, tensile stress in surface layer. The influences of residual stress on the α/β-Sialon SCFGM must be investigated.

Geometric model of α/β-Sialon SCFGM is schematically illustrated in Fig. 3. The model is fully β-Sialon at the center and changes to α-β-Sialon at the two symmetrical surfaces. A commercial finite element code (Algor) was used to analyze the macro thermal residual stresses in the graded structures. In order to simulate the thermal stress by FEM, it was assumed that only elastic deformation would be generated in α/β-Sialon SCFGM. Half of the cross

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section was used for FEM analysis. An axial symmetric boundary condition was applied on the boundaries of r=0. Axial symmetrical element was employed in the computation assuming a reference temperature of 1780℃ and a stress calculation temperature of 25℃. FEM was carried out by using the following values of Young's modulus E, Poisson's ratio ν, thermal expansion coefficient α. E_α=300GPa, ν_α=0.25, $\alpha_\alpha=4.0\times10^{-6}$; E_β=300GPa, ν_β=0.25, $\alpha_\beta=3.0\times10^{-6}$, where the above physical properties were mean values from different literatures. Young's modulus, Poisson's ratios, thermal expansion coefficients and strength of intermediate layers were obtained by the composite mixture rule.

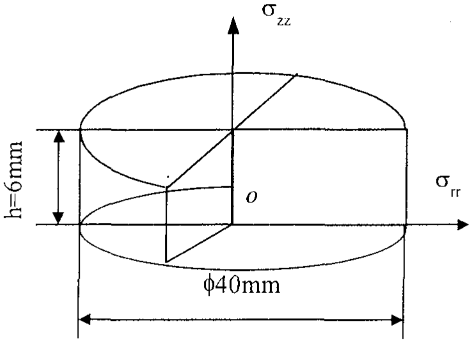

Fig.3 Geometric model of α/β-Sialon SCFGM

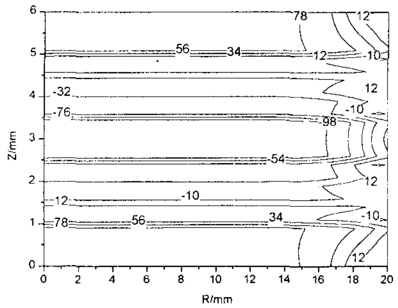

Fig.4 Contour of radial stress of α/β Sialon SCFGM (MPa)

The radial residual stress distribution within α/β-Sialon SCFGM is shown in Fig.4. The stress is concentrated in the interface of graded layers, but all the stresses are too small compared with the compressive strength σ_c

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(3000~4000MPa) and tensile strength σ_t (600~1000MPa) of α/β-Sialon SCFGM, can't lead to the cracking and obvious influence on the mechanical properties of α/β-Sialon SCFGM.

4. Conclusion

(1) The sintering of Sialon ceramics were investigated in the compatible region of α-Sialon and β-Sialon, the relative density of Sialon ceramics increases as the contents of the liquid-phase increases in sintering process.

(2) The α/β-Sialon SCFGM was fabricated by hot pressing, the joint of interface is excellent, no obvious micro-defects and cracks were found.

(3) The residual stresses are too small compared with the compressive strength and tensile strength of α/β-Sialon SCFGM, that will not lead to obvious influence on the mechanical properties of α/β-Sialon SCFGM.

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