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Research Article

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Posted Date: May 13th, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-515032/v1>

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Microwave sintering of duplex α/β -SiAlON ceramic cutting inserts: modifying m , n , z values, sintering temperature and excess Y_2O_3 sintering additive

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Abstract: Duplex α/β -SiAlON ceramic cutting inserts (30α : 70β) were fabricated by microwave sintering. The effects of solid solution parameters (m , n , z), sintering temperature and amount of excess Y_2O_3 sintering additive on phase assemblage, microstructure, mechanical properties and cutting performance were investigated. It was found that increasing m value could improve the formation of α phase while high z value over 1.0 would lead to the dissolution of α phase into β phase and intergranular phase. Increasing amount of excess Y_2O_3 could promote densification and elongated β grain growth, but the excess Y_2O_3 over 4 wt.% would result in substantial crystallization of M'_{ss} phase thus declining the mechanical properties and wear resistance. The microwave-sintered α/β -SiAlON cutting insert with modified parameters ($m = 1.7$, $n = 1.0$, $z = 0.7$ and 3 wt.% excess Y_2O_3) was obtained with optimal comprehensive properties, whose tool life was increased by approximately 75% in high-speed milling of Inconel718 superalloy as compared with commercial α/β -SiAlON cutting insert.

Keywords: α/β -SiAlON; microwave sintering; ceramic cutting inserts; m , n , z value; Y_2O_3 sintering additive

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1 Introduction

SiAlON ceramics are substitutional solid solutions based on Si_3N_4 . Basically, there are two main crystal structures of SiAlON ceramics for engineering applications, namely α -SiAlON and β -SiAlON. The general formula of α - and β -SiAlON are $\text{M}_x\text{Si}_{12-(m+n)}\text{Al}_{(m+n)}\text{O}_n\text{N}_{16-n}$ ($x = m/v$, v means the valence of the modifying cation M (Y_2O_3 , CaO , etc.)) and $\text{Si}_{6-z}\text{Al}_z\text{O}_z\text{N}_{8-z}$ ($0 < z \leq 4.2$) respectively, where m and n values represent Al-N and Al-O substitutions in α - Si_3N_4 crystal lattice and z value represents Al-O substitution in β - Si_3N_4 crystal lattice. In general, α -SiAlON formed as equiaxed grains exhibits higher hardness than β -SiAlON, whereas β -SiAlON formed as elongated hexagonal grains possesses enhanced fracture toughness compared to α -SiAlON [1]. By changing the overall composition, varying $\alpha/(\alpha+\beta)$ ratio duplex α/β -SiAlON ceramic materials with comprehensive properties can be prepared [2].

With well wear resistance and outstanding chemical stability at elevated temperature, duplex α/β -SiAlON ceramics have attracted much interest as cutting tool materials in machining of difficult-to-cut materials like nickel-based superalloys [3]. Researches show the cutting speed of α/β -SiAlON ceramic cutting tools can be up to 300 - 500 m/min in turning nickel-based superalloys and 800 - 1000 m/min when milling [4, 5], which are 10 times than cemented carbide tools. Nevertheless, rapid wear still exists for α/β -SiAlON ceramic cutting tools when high-speed machining nickel-based superalloys [6], and the inefficiency and high cost of SiAlON ceramic cutting tools prepared by the commonly used conventional sintering processes such as pressureless sintering, hot-pressed sintering and hot isostatic pressing sintering limit the development and wide application of SiAlON ceramic cutting tools.

Microwave sintering of SiAlON ceramic tools would be an effective approach to solve the current problems. During microwave sintering process, sample can be directly heated through the interaction with electromagnetic wave that the heating rate can reach 30 - 100 °C/min and the holding time can be

reduced to 0 - 10 min, which are more efficient than conventional sintering (5 - 10 °C/min, 60 - 240 min) [7, 8]. On account of the volumetric fast heating, microwave-sintered ceramics can obtain better densification and mechanical properties [9, 10]. Additionally, as a type of pressureless sintering, microwave sintering has great potential for mass production of complex-shaped cutting inserts whereas pressure sintering methods can not. This is more attractive for low-cost industrial production of ceramic cutting inserts.

In addition to the sintering technology, modifying the overall compositions and sintering conditions could be an effective way to enhance the cutting performance [11]. For α/β -SiAlON ceramics, a deep correlation was discovered among the solid solution parameters, the sintering additives amount, the densification behavior and the microstructural evolution. Based on the literatures [12-16], it can be stated that the m and n values have great impact on the stability of α phase, the z value is an effective parameter on the wear resistance, crystallization of intergranular phases and microstructure, and the sintering additives amount will affect the α -SiAlON phase stability and intergranular phase. Changing the overall compositions of solid solution parameters (m, n, z) and sintering additives will all vary the liquid level and solid solution reaction during sintering process, thus influencing the phase assemblage, microstructure, oxidation resistance, mechanical and machining properties of α/β -SiAlON ceramic cutting inserts [17, 18]. In order to prolong the server life, the optimization of mechanical properties is significant that the higher hardness can improve the resistance to penetration of abrasive particle and thereby reduces the cutting depth whilst the higher fracture toughness reduces the ratio of microcutting to microploughing [19, 20]. The nature and crystallization of secondary phase(s) also play an important part. Acikbas et al. [11] reported α/β -SiAlON cutting tools with crystalline grain boundary ($M'_{SS}, Y_2Si_3-xAl_xO_{3+x}N_{4-x}$) exhibited better cutting performance than that with amorphous phase as the increased

crystallinity and coalescence of the intergranular phase reduced the chemical reaction tendency. In SiAlON literatures, the reported work on microwave-sintered α/β -SiAlON ceramics is few [21], and for cutting tool application is none. Systematically investigation of overall compositions is indispensable for obtaining high quality duplex α/β -SiAlON ceramic cutting inserts by microwave sintering.

In the present work, small batch microwave sintering of duplex α/β -SiAlON ceramic cutting inserts was realized. The effects of solid solution parameters (m, n, z), sintering temperature and amount of excess Y_2O_3 sintering additive were studied. After modifying these parameters, the wear resistance of microwave-sintered α/β -SiAlON ceramic cutting inserts as compared with commercial α/β -SiAlON ceramic cutting insert in dry milling of nickel-based superalloy Inconel718 was studied.

2 Experimental procedure

2.1 Green sample preparation

The composition of the duplex α/β -SiAlON with different m, n, z values was designed by the following formulas:

The mass ratio of α/β was kept constant at 30/70. In addition to the Y_2O_3 dissolved in the α phase, excess Y_2O_3 with amount of 1 - 5 wt.% was added as sintering additive.

Fig. 1(a) displays the preparation process of green sample. The raw powders were composed of α - Si_3N_4 (99.99%, 500 nm, Chaowei, China), α - Al_2O_3 (99.99%, 200 nm, Chaowei, China), AlN (99.99%, 1 μm , Chaowei, China) and Y_2O_3 (99.99%, 50 nm, Chaowei, China). The mixed powders were poured into a Al_2O_3 jar with isopropanol alcohol and Si_3N_4 balls, and then ball milled in a planetary ball mill (Model QM-3SP2, Nanjing, China) for 2 h at a speed of 250 rpm. The ball-to-powder mass ratio was 8:1. 2 wt.%

PVA was added in the slurries as binder. After ball milling, the slurries were dried in a vacuum drying oven, ground by agate grinding bowl and sieved through a 100-mesh. Finally, green samples were compacted into a cylinder with the diameter of 16 mm under uniaxial pressure of 150 MPa.

Fig. 1. (a) Preparation process of green sample, (b) diagrammatic sketch of the microwave insulation box, (c) diagrammatic sketch of the microwave sintering system, (d) microwave-sintered α/β -SiAlON cutting inserts.

2.2 Microwave sintering

Fig. 1(b) displays the diagrammatic sketch of the microwave insulation box. BN crucible was placed in the center of the mullite insulation box, covered with mullite insulation cotton. Three green samples were placed flat in the center of BN crucible, covered with SiC susceptor to help with the sintering. Fig. 1(c) displays the diagrammatic sketch of the microwave sintering system. The heat insulation box was put on the base of a microwave atmosphere furnace (ATPIO XO-5kW, China) with a rotation speed of 5 r/min. Through the thermometer pore of the microwave insulation box, the real time temperature of the samples can be measured by the infrared thermometer. Before sintering, air in the furnace chamber would be pumped out and replaced by N₂. In the initial stage of sintering process, the microwave atmosphere

furnace operated at minimum power of 900 W during which the heating rate was about 50 - 100 °C/min.

Then, the heating rate was kept constant at 30 °C/min between 470 - 1400 °C and 15 °C/min after 1400 °C until reached the desired temperature (1690 - 1750 °C). The holding time was 10 min. After sintering, samples were naturally cooled to room temperature in the furnace. The microwave-sintered α/β -SiAlON cutting inserts are shown in Fig. 1(d).

2.3 Characterization

The bulk density of the sintered samples was calculated by the Archimedes' method. The Vickers hardness was measured through indentation method under a load of 98 N and holding time of 15 s. Fracture toughness measurement was based upon the Vickers indentation method. The formula of the fracture toughness of the samples is as follows [22]:

$$K_{IC} = 0.203 * (c/a)^{\frac{3}{2}} * \sqrt{a} * HV \quad (3)$$

where K_{IC} is the fracture toughness ($\text{MPa} \cdot \text{m}^{1/2}$), a and c are half the length of the indentation diagonal (mm) and the crack (mm) respectively ($c/a > 2.5$), and HV is the Vickers hardness (GPa). The microstructure of the polished surface of the sintered samples was observed by Scanning electron microscopy (SEM, Hitachi S-4800, Japan) by using back-scattered electron imaging mode.

X-ray diffraction analyses (XRD, Bruker-AXS D8 Advance, USA) was conducted to detect the $\alpha/(\alpha+\beta)$ ratio and the types of other phases. The $\alpha/(\alpha+\beta)$ ratio was calculated from the XRD patterns by the following formula:

$$\frac{I_{\alpha}}{I_{\alpha} + I_{\beta}} = \frac{1}{1 + K [1/(1 - \omega_{\alpha}) - 1]} \quad (4)$$

where ω_{α} is mass ratio of $\alpha/(\alpha+\beta)$, I_{α} is the intensity of the stronger peak of α (1 0 2) and α (2 1 0), I_{β} is the intensity of the stronger peak of β (1 0 1) and β (2 1 0), and K is the combined proportionality constant calculated by the equation: $K = K_{\alpha}/K_{\beta}$, where K_{α} is 0.518 for α (1 0 2) reflection and 0.544 for

α (2 1 0) reflection, and K_β is 0.518 for β (1 0 1) reflection and 0.544 for β (2 1 0) reflection [23].

2.4 Machining test

Fig. 2 shows the schematic diagram of the machining test. The wear resistance of microwave-sintered duplex α/β -SiAlON ceramic milling inserts and commercial duplex α/β -SiAlON ceramic milling insert (Kennametal KYS30, RNGN120400EGN) was evaluated in a machining test by rough milling of Inconel718 superalloy ($v_c = 800$ m/min, $a_p = 1.5$ mm, $f_z = 0.12$ mm/z). The flank wear was measured by a digital microscope (Insize ISM-PM200S, China) during testing in intervals of 1 cut (cutting length: 150 mm, width: 30 mm). The diameter and thickness of the round insert are 12.7 mm and 4.7 mm respectively, which was clamped on one tooth of Kennametal KCRA63Z06S22RN12 indexable face mill. The tool parameters of the face mill were shown in Fig. 2(b).

Fig.2. (a) Schematic diagram of machining test, (b) tool parameter.

3 Results and discussion

3.1 Orthogonal experiment (m , n , z values and sintering temperature)

Orthogonal experiment was carried out firstly for selecting the general scope of appropriate m , n , z values and sintering temperature. The primary purpose was to obtain α/β -SiAlON ceramics with designed composition ($\alpha/(\alpha+\beta)$ ratio = 30%, without residual raw powder phase) and the second purpose was to screen the influencing parameters for obtaining well mechanical properties (hardness and fracture

toughness). The detailed experimental design and results are given in Table 1, and the corresponding XRD pattern is shown in Fig. 3. The m and n values of α -SiAlON were set at 1.0, 1.35 and 1.7 respectively, the z value of β -SiAlON was set at 0.4, 0.7 and 1.0 respectively, and the sintering temperature was set at 1690°C, 1720°C and 1750°C respectively. 2 wt.% excess Y_2O_3 was added as sintering additive.

From Table 1, the detected $\alpha/(\alpha+\beta)$ ratio (wt.%) was calculated between 2 - 29% ($< 30\%$), in which some α phase were converted to β phase. Moreover, residual α - Si_3N_4 phase was detected in sample S1, S5, S6 and S9, in which the intensity in S1 was relatively strong (Fig. 3). This indicated the solid solution reaction was not complete, which could be resulted from the low sintering temperature (1690 - 1720°C) or low liquid content of these components [12]. For the samples sintered at 1750°C (S3, S4 and S8), α - Si_3N_4 was dissolved, and higher bulk density over $3.22 \text{ g}\cdot\text{cm}^3$ was obtained. For the secondary phases, M'_{SS} phase (melilite phase = $Y_2Si_{3-x}Al_xO_{3+x}N_{4-x}$) was detected in all samples as intergranular phase, while 21R phase (AlN polypoid = $Si_3Al_4N_8$) was detected in sample S3, S5 and S7 with high z value of 1.0. For cutting tool application, M'_{SS} phase could be a better phase for SiAlON ceramics as it displayed excellent high temperature stability due to its very high nitrogen content [24].

Table 1. Orthogonal design and corresponding bulk density, phase assemblage and mechanical properties of the α/β -SiAlON ceramics

Sam- ple	Orthogonal design				Bulk	Phase assemblage				HV10	K _{IC}
	<i>m</i>	<i>n</i>	<i>z</i>	Sintering	Density	<i>α</i> /	Other phases			/GPa	/MPa • m ^{1/2}
	value	value	value	temperature	/g•cm ³	(α+β)	M' _{SS}	21R	α-		
	(A)	(B)	(C)	/°C (D)		ratio			Si ₃ N ₄		
S1	1 (1.0)	1 (1.0)	1 (0.4)	1 (1690)	3.10	11%	w		m	13.65 ± 0.15	6.24 ± 0.23
S2	1 (1.0)	2 (1.35)	2 (0.7)	2 (1720)	3.20	12%	w			15.46 ± 0.19	5.50 ± 0.28
S3	1 (1.0)	3 (1.7)	3 (1.0)	3 (1750)	3.22	2%	w	w		15.22 ± 0.15	5.99 ± 0.23
S4	2 (1.35)	1 (1.0)	2 (0.7)	3 (1750)	3.23	18%	m			15.90 ± 0.14	5.27 ± 0.12
S5	2 (1.35)	2 (1.35)	3 (1.0)	1 (1690)	3.20	13%	w	w	w	15.09 ± 0.17	5.31 ± 0.18
S6	2 (1.35)	3 (1.7)	1 (0.4)	2 (1720)	3.16	20%	w		vw	15.21 ± 0.47	5.27 ± 0.25
S7	3 (1.7)	1 (1.0)	3 (1.0)	2 (1720)	3.23	21%	w	w		15.78 ± 0.13	5.52 ± 0.08
S8	3 (1.7)	2 (1.35)	1 (0.4)	3 (1750)	3.24	27%	w			16.38 ± 0.07	5.71 ± 0.30

S9	3 (1.7)	3 (1.7)	2 (0.7)	1 (1690)	3.19	29%	w	vw	15.91 ± 0.16	5.54 ± 0.22
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m = medium, w = weak, vw = very weak.

Fig. 3. XRD pattern of the orthogonal designed α/β -SiAlON ceramics (M'_{SS} : mellite phase = Y_2Si_3 -

Table 2 shows the range analysis results of $\alpha/(\alpha+\beta)$ ratio, hardness and fracture toughness of the orthogonal experiment. Taking $\alpha/(\alpha+\beta)$ ratio as the index, the factor level of m value (A) was much greater than other factors. Along with the increasing m value from 1.0 to 1.7, the average result of $\alpha/(\alpha+\beta)$ ratio was evidently promoted from 8.3% to 25.7%, which was closer to the setting value of 30%. This phenomenon could be connected to the increasing amount of Y_2O_3 in the component ($x = m/3$, formula (1)). With higher amount of Y_2O_3 , more rare earth cations could be incorporated into the α -SiAlON structure, thus improving the formation and stability of α phase [15]. On the other hand, the variations of other parameters with the average result of $\alpha/(\alpha+\beta)$ ratio were not obvious. Only when z value increased to 1.0 did a decline from 19.7% to 12.0% occurred. This might be attributed to the excessive oxygen content induced by the high z value, which damaged the stability of α -SiAlON phase [25]. For obtaining high $\alpha/(\alpha+\beta)$ ratio ceramics, low z value of 0.4 - 0.7 was appropriate.

Taking hardness as the index, the factor levels of m , n , z values and sintering temperature were 1.25, 0.53, 0.67 and 0.95 respectively. It meant m value and sintering temperature played more important parts.

With the increasing of m value and sintering temperature, the average results of hardness were enhanced. The amelioration of hardness induced by m value could be attributed to the increasing of $\alpha/(\alpha+\beta)$ ratio, whereas the other induced by sintering temperature could be resulted from the improvement of densification. Taking fracture toughness as the index, it can be observed the factor level of m value (A) was greater than other factors, but it was still small ($0.63 \text{ MPa}\cdot\text{m}^{1/2}$) and the variations were irregular for all factors. This indicated that m , n , z values and sintering temperature had no significant impact on the fracture toughness within the setting range. According to these results, high m value of 1.7 was required for improving the formation of α phase and sintering temperature of 1750°C was necessary for enhancing the densification and mechanical properties. The impacts of n and z values were not evident, which still needed to be further studied.

Table 2. Range analysis results of hardness, fracture toughness and $\alpha/(\alpha+\beta)$ ratio of the orthogonal experiment

	$\alpha/(\alpha+\beta)$ ratio				HV10/GPa				$K_{IC}/\text{MPa}\cdot\text{m}^{1/2}$			
	A	B	C	D	A	B	C	D	A	B	C	D
$\bar{k}_{1j}$	8.3%	16.7%	19.3%	17.6%	14.78	15.11	15.08	14.88	5.91	5.68	5.74	5.70
$\bar{k}_{2j}$	17.0%	17.3%	19.7%	17.7%	15.40	15.64	15.75	15.48	5.28	5.51	5.43	5.43
$\bar{k}_{3j}$	25.7%	17.0%	12.0%	15.7%	16.02	15.45	15.36	15.83	5.56	5.60	5.61	5.66
R	17.4%	0.6%	7.7%	2.0%	1.25	0.53	0.67	0.95	0.63	0.17	0.31	0.27

Note: $\bar{k}_{ij}$ means the average result of each factor at each level in Table 1, and R means the factor level of each factor.

3.2 Effects of z value on phase assemblage, microstructure and mechanical properties of α/β -SiAlON ceramics

In α/β -SiAlON ceramics, n and z values represent Al-O substitution in Si-N of α - and β -SiAlON respectively. When $\alpha/(\alpha+\beta)$ ratio is 30%, the effect of increasing n value by 0.7 in the component is consistent with the effect of increasing z value by 0.14 (for example, the component of the Y- α/β -SiAlON with n value of 1.7 and z value of 0.4 is equal to that with n value of 1.0 and z value of 0.54 while m

value is the same). As a result, it can be inferred that the impact of n and z values on α/β -SiAlON ceramics is similar in a certain range. Since β ratio was higher in this research, the effects of z value were studied, and the z value was set at 0.4, 0.54, 0.7, 0.84, 1.0 and 1.14 respectively. The m and n values were kept constant at 1.7 and 1.0 respectively, and the excess Y_2O_3 was 2 wt.%. The sintering temperature was set at 1750°C.

Fig. 4(a) shows the XRD pattern of the α/β -SiAlON ceramics with different z value. Weak α - Si_3N_4 peaks were detected in the sample with z value of 0.4 since the oxygen content was low, and disappeared as z value increased. For the secondary phases, M'_{ss} phases were detected in all samples and 21R phases were detected as z value exceeded 1.0 (same as the orthogonal experiment result in Fig. 3). In addition, substantial crystallization was observed as z value increased to 1.14, which could be resulted from the high oxygen content. Fig. 4(b) shows the $\alpha/(\alpha+\beta)$ ratio variation with z value. Under z value of 0.4 to 0.84, the $\alpha/(\alpha+\beta)$ ratio varied between a narrow range of 27% - 36% (close to setting value of 30%). When z value increased to 1.0 and 1.14, the $\alpha/(\alpha+\beta)$ ratio evidently decreased to 15% and 14% respectively. Correspondingly seen from the XRD patterns in Fig. 4(a), the intensities of α -SiAlON phase (peak at 34.0° and 35.0° 2 θ) were also obviously reduced.

Reported by Carmen et al. [26], there was a reversible thermodynamic transformation relationship between α - and β -SiAlON phases: $\alpha + \text{liquid} \leftrightarrow \beta + M'_{ss} + 21R$. In his research, long time heat-treatment at 1450°C for 72 h led to $\alpha \rightarrow \beta$ transformation and the reverse $\beta \rightarrow \alpha$ transformation was observed under subsequent heat treatment at 1800°C for 4 h, accompanied with the change of M'_{ss} phase and 21R phase. The observation in this research when z value exceeded 1.0 was same to the $\alpha \rightarrow \beta$ transformation reaction that the $\alpha/(\alpha+\beta)$ ratio was reduced and the crystallization of M'_{ss} and 21R phases was enhanced. In α/β -SiAlON ceramics, the stability of the α phase is connected to the liquid phase and temperature and

will dissolve while the conditions appear to permit significant diffusion [27]. When z value was 1.0, the liquid content exceeds critical level, and this results in the dissolution of α phase. Acikbas et al. has also studied the effect of z value (0.3 - 1.1) on phase evolution of α/β -SiAlON prepared at 1840 - 1890°C but the $\alpha/(\alpha+\beta)$ ratio was barely changed and no 21R phase was detected [18]. This observation was different, but it was still in agreement with our explanation that the α -SiAlON phase was more stable at high temperature according to the $\beta \rightarrow \alpha$ transformation.

Fig. 4. (a) XRD pattern (M'_{SS}: melilite phase = Y₂Si_{3-x}Al_xO_{3+x}N_{4-x}; 21R: AlN polypoid = Si₃Al₄N₈), and

(b) $\alpha/(\alpha+\beta)$ ratio, of the α/β -SiAlON ceramicss with different z value ($m = 1.7$, $n = 1.0$, sintered at 1750°C).

Fig. 5 shows the microstructures of the α/β -SiAlON ceramics with different z values taken by SEM-BSE mode. The microstructure presented three different contrasting phases, which were α -SiAlON (light grey), β -SiAlON (dark grey) and intergranular phase (white area). It can be observed the α -SiAlON phase presented as equiaxed grains, and the grain size was very similar in all materials. The β -SiAlON phase mainly presented as prismatic grains, and the grain size was relatively coarsened with the increasing of z value (Fig. 5(e & f)). Under high z value of 1.0 and 1.14, the area of β -SiAlON was obviously facilitated, which was consistent with the variation of $\alpha/(\alpha+\beta)$ ratio in Fig. 4. Moreover, the intergranular phase was

generally distributed around the SiAlON grains. In the sample with higher z value, there should be more aluminium in intergranular phase that the Si-N bonds in M'_{ss} phase ($Y_2Si_{3-x}Al_xO_{3+x}N_{4-x}$) can be progressively replaced by Al-O bonds. This can lead to the increase of oxygen content, which could in turn improve the oxidation resistance of the material [2, 28].

Fig. 5. Microstructure of α/β -SiAlON ceramics with different z values ($m = 1.7$, $n = 1.0$, sintered at $1750^\circ C$) taken by SEM-BSE mode, (a) $z = 0.4$, (b) $z = 0.54$, (c) $z = 0.7$, (d) $z = 0.84$, (e) $z = 1.0$, (f) $z =$

Fig. 6 shows the bulk density, hardness and fracture toughness variations with z value. The bulk density measured in the range of 3.23 - 3.24 g·cm³ was close to the theoretical density based on the literatures [17, 29]. The hardness and fracture toughness of the α/β -SiAlON ceramics had an overall declining trend with the increase of z value, but the change was not significant. As z value increased from 0.4 to 0.84, the mechanical properties of the samples narrowly varied in a small range (hardness: 16.35 - 16.76 GPa, fracture toughness: 5.35 - 6.15 MPa·m^{1/2}) since they have similar $\alpha/(\alpha+\beta)$ ratio (29% - 36%). When z value exceeded 1.0, the hardness was declined to 16.02 $\pm$ 0.12 GPa due to the dissolution of α phase (Fig. 4(b)). And when z value increased to 1.14, both hardness and fracture toughness were declined to 15.46 $\pm$ 0.07 GPa and 4.91 $\pm$ 0.09 MPa·m^{1/2} respectively. This could be attributed to the substantial crystallization of the intergranular phases (Fig. 4) since their mechanical properties were relatively poor (hardness: 12 - 15 GPa, fracture toughness: 4 - 5 MPa·m^{1/2} [30]). Although the α/β -SiAlON ceramic with low z value possessed better mechanical properties, it was reported SiAlON ceramics with low z value (e.g. 0.2 - 0.3) exhibited a short tool life in machining of superalloy due to chemical wear [31]. Besides, α/β -SiAlON ceramic with low z value was also hard for sintering as residual α -Si₃N₄ was detected (Fig. 4(a)). Thus, the z value of 0.54 - 0.84 could be appropriate for microwave sintering of α/β -SiAlON ceramics.

Fig. 6. Bulk density, hardness and fracture toughness variations with z value.

3.3 Effects of the amount of excess Y_2O_3 sintering additive on phase assemblage, microstructure, mechanical properties and cutting performance of α/β -SiAlON ceramic milling inserts

The effect of the amount of excess Y_2O_3 sintering additive was investigated. From the above, the m , n , z values were set at 1.7, 1.0 and 0.7 respectively, and the sintering temperature was set at 1750°C. α/β -SiAlON ceramics with the amount of excess Y_2O_3 of 1 - 5 wt.% were fabricated, and named Y1 - Y5.

Fig. 7 shows the XRD pattern of the sample Y1 - Y5 and $\alpha/(\alpha+\beta)$ ratio variation with the amount of excess Y_2O_3 . Seen from the XRD patterns, M'_{ss} phase was detected as the only intergranular phase in all the materials. The intensity of M'_{ss} phase (peak at 32.3° 2θ , colored area in Fig. 7(a)) was relatively weak in sample Y1, Y2 and Y3, and significantly enhanced when the amount of excess Y_2O_3 exceeded 4 wt.%. On the other hand, the $\alpha/(\alpha+\beta)$ ratio was measured at 31%, 29%, 34% and 28% respectively for the sample Y1, Y2, Y3 and Y5 (close to the setting value of 30%). Only when the amount of excess Y_2O_3 was 4 wt.% did the $\alpha/(\alpha+\beta)$ ratio declined to 20%. In high temperature sintering of α/β -SiAlON ceramics, there should be two destinations for Y_2O_3 : incorporated into α -SiAlON structure or intergranular phase. As we discussed in section 3.1, the increasing amount of Y_2O_3 induced by the growing m value remarkably promoted the $\alpha/(\alpha+\beta)$ ratio to the setting value (Table 2) whereas the crystallization level of intergranular phase was low (Table 1). It could be inferred that Y_2O_3 preferentially tended to incorporated into α -SiAlON, and the sample Y1 - Y3 might have higher x value than designed value since their $\alpha/(\alpha+\beta)$ ratios had reached 30% after modifying the m value to 1.7. For sample Y4, the Y_2O_3 amount reached a critical level and substantial crystallization of M'_{ss} phase was observed (Fig. 7(a)). After the devitrification of M'_{ss} phase, the available rare earth cations in the liquid were consumed, hence the α/β -

SiAlON shifted to β -SiAlON. As the amount of excess Y_2O_3 continued to increase to 5 wt.%, the $\alpha/(\alpha+\beta)$ ratio increased again to the setting value.

Fig. 7. (a) XRD pattern (M'_{ss} : melilite phase = $Y_2Si_{3-x}Al_xO_{3+x}N_{4-x/8}$), and (b) $\alpha/(\alpha+\beta)$ ratio, of the α/β -SiAlON ceramics with different amount of excess Y_2O_3 ($m = 1.7$, $n = 1.0$, $z = 0.7$, sintered at $1750^\circ C$).

Fig. 8 shows the microstructures of α/β -SiAlON ceramics with different amount of excess Y_2O_3 sintering additive. The α -SiAlON phase presented as equiaxed grains (light grey) and had similar grain size in all materials. The β -SiAlON grains (dark grey) were relatively finer in sample Y1 and many β -SiAlON grains appeared to be equiaxed with low aspect ratio (Fig. 8(a)). For comparison, more elongated β -SiAlON grains were observed in sample Y2 - Y5 and the β -SiAlON grains were relative coarsened in sample Y4 and Y5. This was in agreement with Herrmann et al. [13] that the rare-earth-containing liquid phase had decisive role for elongated grain growth at high sintering temperature. In addition, the intergranular phase (white area) was distributed around SiAlON grains. In accordance with the increase of excess Y_2O_3 , more white area of the rare-earth-rich melilite phase was observed (Fig. 8(d & e)), illustrating the facilitating of intergranular phase.

Fig. 8. Microstructure of α/β -SiAlON ceramics with different amount of excess Y_2O_3 sintering additives ($m = 1.7$, $n = 1.0$, $z = 0.7$, sintered at 1750°C) taken by SEM-BSE mode, sample (a) Y1, (b) Y2, (c) Y3, (d) Y4, (e) Y5.

Fig. 9 shows the bulk density, hardness and fracture toughness variations with the amount of excess Y_2O_3 . The bulk density of the α/β -SiAlON ceramics was gradually increased from 3.23 to $3.29 \text{ g}\cdot\text{cm}^{-3}$ with the increase of excess Y_2O_3 . This could be mainly due to the high density of Y_2O_3 ($5.01 \text{ g}\cdot\text{cm}^{-3}$) and moderate improvement of densification though near full stage was reached. In addition, the hardness was

barely changed as the amount of excess Y_2O_3 increased from 1 wt.% to 3 wt.% (16.48 - 16.59 GPa) since the content of α phase was similar in sample Y1 -Y3 (29% - 34%), whereas the fracture toughness was gradually enhanced from 5.19 ± 0.13 MPa·m^{1/2} to 5.82 ± 0.30 MPa·m^{1/2} due to the elongated β grain growth (Fig. 8). When the amount of excess Y_2O_3 increased to 4 wt.% and 5 wt.%, owing to the substantial crystallization of M'_{ss} phase, the hardness was evidently reduced to 15.71 ± 0.12 GPa and 15.62 ± 0.08 GPa respectively and fracture toughness was declined to 4.77 ± 0.14 MPa·m^{1/2} 4.64 ± 0.22 MPa·m^{1/2} respectively.

Fig. 9. Bulk density, hardness and fracture toughness variations with the amount of excess Y_2O_3 .

Fig. 10 shows the tool life of microwave-sintered α/β -SiAlON ceramic cutting inserts with different amount of excess Y_2O_3 and commercial α/β -SiAlON ceramic cutting insert (KYS30) in rough milling of Inconel718 superalloy. It can be observed that the tool life of microwave-sintered α/β -SiAlON ceramic cutting inserts was increased as the amount of excess Y_2O_3 increased from 1 wt.% to 3 wt.% and declined when it exceeded 4 wt.%. Sample Y1 was obtained with the shortest tool life since its density was relatively poor (Fig. 9) and the aspect ratio of β grains was small (Fig. 8(a)). With the enhanced densification, elongated β grains and mechanical properties induced by the increasing amount of excess Y_2O_3 , the tool life was well improved for sample Y2 and Y3. Moreover, the tool life of sample Y4 and

Y5 was declined but not expected catastrophic (better than Y1 and close to KYS30) although their mechanical properties were poor (Fig. 9). This was probably owing to the increased crystallinity and coalescence of the intergranular phase that reduced the chemical reaction tendency of α/β -SiAlON cutting inserts [19] and the elongated β grain growth resulted from the high excess Y_2O_3 amount (Fig. 8).

Sample Y3 was obtained with the optimal wear resistance, and its flank wear morphologies were displayed in Fig. 10 as compared with commercial α/β -SiAlON ceramic cutting insert (KYS30). No spalling was observed in both inserts, and the sample Y3 exhibited better wear resistance, whose tool life was increased by approximately 75% than KYS30. Table 3 shows the comparison of microwave-sintered and commercial α/β -SiAlON milling inserts. It can be seen KYS30 was measured with lower $\alpha/(\alpha+\beta)$ ratio (18%) than sample Y3 (34%) so that KYS30 possessed lower hardness and higher fracture toughness, but the difference was slight. Besides, the intergranular phase of KYS30 was detected as amorphous, while sample Y3 was detected as M'_{ss} . The crystallization of M'_{ss} could improve the wear resistance of SiAlON cutting inserts, but there might exist other factors affecting the cutting performance since the sintering technology and composition (cation type, solid solution parameters, sintering additives) of KYS30 was not available. This comparison can indicate microwave sintering of high quality α/β -SiAlON ceramic cutting inserts was feasible was valuable.

Fig. 10. Tool life of microwave-sintered α/β -SiAlON ceramic cutting inserts with different amount of excess Y_2O_3 and commercial α/β -SiAlON ceramic cutting insert.

Table 3. Comparison of microwave-sintered and commercial α/β -SiAlON milling inserts

Sample	Bulk density /g·cm ³	$\alpha/(\alpha+\beta)$ ratio /(wt.%)	Intergranular phase	Cation type	Hardness /GPa	Fracture toughness /MPa·m ^{1/2}
Kennametal KYS30	3.33	18 %	Amorphous	-	16.30 ± 0.38	6.15 ± 0.14
Y3	3.25	34 %	M' _{ss} (w)	Yttrium	16.48 ± 0.04	5.82 ± 0.30

w = weak.

4 Conclusions

Duplex α/β -SiAlON ceramic cutting inserts with nominal composition of $\alpha: \beta = 30: 70$ were successfully fabricated by microwave sintering. The solid parameters (m, n, z) and sintering temperature were optimized by orthogonal experiment, and the effects of z value and amount of excess Y_2O_3 sintering additive on phase assemblage, microstructure, mechanical properties and wear resistance were systematically investigated. The main conclusions are as follows:

- The increasing m value from 1.0 to 1.7 could promote the formation of α phase (8.3% to 25.7%) thus improving the hardness, and the high sintering temperature could promote the solid solution reaction, densification and hardness.
- $\alpha + \text{liquid} \rightarrow \beta + M'_{ss} + 21R$ transformation reaction was observed when z value exceeded 1.0 under sintering temperature of 1750°C. Observed from microstructure, the area of β -SiAlON phase was noticeably increased when z value exceeded 1.0, and the β grain growth was relatively accelerated with the increasing z value. The hardness and fracture toughness of α/β -SiAlON ceramics had an overall declining trend with the increasing z value, and the sample with z value of 1.14 was obtained with worst mechanical properties due to the substantial crystallization of intergranular phase.
- Increasing excess Y_2O_3 amount could improve the grain growth and aspect ratio of the β -SiAlON

grains and enhance the densification. But the excess Y_2O_3 over 4 wt.% would lead to the substantial crystallization of M'_{ss} phase and decline the mechanical properties and wear resistance of α/β -SiAlON ceramics.

- The microwave-sintered α/β -SiAlON cutting insert with modified parameters (m , n , z value and excess Y_2O_3 of 1.7, 1.0, 0.7 and 3 wt.% respectively, sintered at 1750°C) was prepared with the optimal comprehensive properties. Compared with commercial α/β -SiAlON cutting insert, the tool life was increased by approximately 75% in rough milling of Inconel718 superalloy.

Acknowledgements

The work is supported by National Natural Science Foundation of China (52075266, 51875291), Excellent Youth Fund of Jiangsu Province (BK20190070) and Jiangsu Provincial Six Talent Peaks Project (GDZB-016).

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