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Correlation of wear behavior and indentation fracture resistance in silicon nitride ceramics hot-pressed with alumina and yttria

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Abstract

Nine kinds of silicon nitrides with different microstructures were fabricated by controlling both sintering conditions and amounts of sintering additives, Al_2O_3 and Y_2O_3. The wear behavior of the various Si_3N_4 ceramics was investigated in sliding contact test without lubricant. The specific wear rate varied notably from 6×10^{-6} to 4×10^{-4} $mm^3\ N^{-1}\ m^{-1}$ depending on the microstructures, whose ranking was difficult to predict directly from the hardness or fracture resistance obtained by the indentation fracture (IF) technique as well as the single-edge-precracked beam (SEPB) method. A good correlation was obtained between the specific wear rate and both mechanical properties when a lateral-crack chipping model was applied as the material removal process. However, the correlation was lost when the fracture toughness obtained by the SEPB method was employed, indicating that the conventional long-crack toughness is inappropriate for analyzing the wear behavior of Si_3N_4 exhibiting a rising *R*-curve behavior.

Keywords: Si_3N_4; Wear resistance; Toughness and toughening; Hardness; Indentation fracture technique

1. Introduction

Silicon nitride is an attractive material for tribological applications since it possesses superior wear resistance and advantages such as light weight, high strength and toughness, good corrosion resistance and a low thermal expansion coefficient. Many studies have been conducted to improve its mechanical properties through microstructural control, such as "self-reinforcing" mechanism. Although there has been much research on the tribological behavior of silicon nitride, many of them have been focused on the relation between the sliding environment and the wear resistance,[1–9] and relatively fewer studies have explored the effect of microstructure on the wear performance.[4,10–13] However, the reported relationships between microstructure and tribological performance are often contradictory, leaving fundamental understanding of its wear mechanism to be clarified. For example, Zutshi et al., Gomes et al. and Doğan and Hawk found that the wear resistance was improved when the grain size became smaller despite of a decrease in fracture toughness.[10–12] By contrast, Carrasquero et al. demonstrated that the "self-reinforced" microstructure with elongated grains suppressed the propagation of cracks, which not only improved both fracture toughness and strength but also enhanced the wear resistance.[13] In the case of silicon carbide whisker-reinforced silicon nitride composites, Wang and Mao reported that the pullout mechanism of SiC whisker made the composite more wear resistant than the monolithic silicon nitride,[14] while Gomes et al. and Doğan and Hawk observed little difference between the wear performances for them.[15,16] In our previous studies, the influence of microstructure on the friction and wear properties of various Si_3N_4 ceramics was investigated in sliding contact test without lubricant.[17,18] It was found that the tribological properties varied considerably depending on the microstructures. However, a clear relationship between the tribological characteristics and the grain size as well as the mechanical properties was not obtained.

The complicated results from all these researches may be attributable to the complexity of the experimental conditions, especially to the diversity of the samples tested in these literatures, that is, the chemical composition and crystallinity of the intergranular phase as well as the ratio of α/β phase of silicon

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nitride were different besides variations in the grain size distribution and morphology.[11,12] In this study, in order to elucidate the effect of the grain size and morphology on the tribological property, nine silicon nitride samples were hot-pressed using equal amounts of alumina and yttria as sintering additives to yield an intergranular amorphous phase with almost the same chemical composition, while the distribution of grain size was controlled by both sintering conditions and total amounts of additives. The samples were tested in unlubricated sliding test at an ambient atmosphere to correlate the volume wear loss to both microstructures and mechanical properties such as hardness and fracture toughness. An indentation fracture model for material removal process in abrasive wear, which was developed by Evans and Marshall,[19] was employed to explain the dependence of the wear volume on an inverse function of the hardness and fracture toughness.

2. Experimental procedure

2.1. Materials

Two sets of silicon nitride ceramics (listed in Table 1) were fabricated from Si_3N_4 powder with Y_2O_3 and Al_2O_3 as sintering additives. The amounts of the sintering additives of the first set of samples (A series) were fixed at 5 wt% Al_2O_3/5 wt% Y_2O_3. In order to alter both grain size distributions and grain morphologies of the silicon nitride samples, the combination of the sintering temperature and soaking time was varied as follows; 1750 °C for 1 h, 1850 °C for 1 h, 1950 °C for 2 h and 1950 °C for 8 h. By contrast, the amount of additives of the second set of samples was increased progressively from 1 wt% Al_2O_3/1 wt% Y_2O_3 to 10 wt% Al_2O_3/10 wt% Y_2O_3 while the sintering condition was set constant at 1950 °C for 2 h (B series). The ratios of Y_2O_3 to Al_2O_3 in weight percent were kept at unity for all compositions, so that the chemical compositions of intergranular phases were considered to be almost identical. However, the content of silica from the silica on the surface of raw Si_3N_4 powder was supposed to decrease slightly with an increase in the additives. The percentage of nitrogen in the glass phase might also vary depending on the compositions. The starting powders were mixed in ethanol for 24 h. The slurry was dried and passed through a 150-mesh sieve. The mixed powders were hot-pressed in a graphite furnace at temperatures ranging from 1750 to 1950 °C under a pressure of 30 MPa in a nitrogen atmosphere. The detailed fabrication procedure was described in our previous reports.[17,18] The densities of the as-sintered bodies were measured using the Archimedes technique. The crystal phases in the specimens were identified by X-ray diffraction (XRD). The machined samples were polished and plasma etched in CF_4 gas before microstructural observation by scanning electron microscopy (SEM). The diameter of each grain was evaluated by the shortest grain diagonal in two-dimensional images with magnifications of 10k and/or 5k. In order to count coarse and elongated grains (major axis $\gtrsim$8 μm) which occasionally appeared in the samples sintered at higher temperatures, the micrographs with the lower magnification of 1k were also used. Aspect ratios of the normal grains were estimated from the mean value of the 10% highest observed aspect ratios.[20] Aspect ratios of the coarse grains selected from the micrographs at a magnification of 1k were calculated by the same procedure as mentioned above.

2.2. Test procedure

Young's modulus was measured by the ultrasonic pulse echo method as used in Japanese industrial standard (JIS) R 1602.[21] Vickers indentations were made on the polished surface perpendicular to the hot-pressing axis with a hardness tester (Model AVK-C2, Akashi Corp., Yokohama, Japan). Both Vickers hardness and indentation fracture resistance were evaluated at a load of 294 N with the dwell time of 15 s. Eight impressions were made for each sample. The lengths of the impression diagonals, $2a$, and sizes of surface cracks, $2c$, were measured with a traveling microscope (Model MM-40, Nikon Corp., Tokyo, Japan) immediately after the unloading. A 10× eyepiece and a 50× objective were used to observe the bright field images of the indentations. The fracture resistance, K_R, was calculated using the Miyoshi's equation as follows,[22,23] since it was reported that

Table 1
Composition of the starting powders, sintering conditions, median grain diameter and aspect ratio of the silicon nitride samples hot-pressed with alumina and yttria as sintering additives

Material designation	Amount of sintering additives (wt%)		Sintering condition		Median grain diameter (μm)	Aspect ratio[a]	
	Al_2O_3	Y_2O_3	Temperature (°C)	Soak time (h)		Normal grain	Coarse grain
A1	5.0	5.0	1750	1	0.23	4.5	–
A2	5.0	5.0	1850	1	0.28	4.9	–
A3	5.0	5.0	1950	2	0.52	4.9	12.0
A4	5.0	5.0	1950	8	0.91	5.5	9.3
B1	1.0	1.0	1950	2	0.35	4.1	5.2
B2	1.75	1.75	1950	2	0.34	4.7	5.6
B3	2.5	2.5	1950	2	0.40	4.7	10.4
B4	3.34	3.34	1950	2	0.32	4.5	10.4
B5	10.0	10.0	1950	2	0.59	5.9	11.3

[a] The aspect ratios of the normal grains were obtained from the 5k and 10k magnification images, whereas those of coarse grains were calculated using the selected grains with major axis >8 μm in micrographs of 1k magnification.