MATERIAL DESIGN FOR HIGH-PERFORMANCE SILICON NITRIDE CERAMICS

MAMORU MITOMO
National Institute for Research in Inorganic Materials,
1-1, Namiki, Tsukuba-shi, Ibaraki, 305, Japan

ABSTRACT

Microstructure design for high-performance silicon nitride ceramics is shown. High strength materials should have fine and uniform microstructures. Bimodal microstructures (in-situ composites) are expected to give high fracture toughness. In-situ composites, which show R-curve behavior, have been fabricated from β powder with wide grain size distribution. The mechanical properties of in-situ composites are discussed in relation to the size and distribution of large grains.

INTRODUCTION

The improvement of mechanical properties of silicon nitride ceramics has been tried by optimizing powder characteristics or fabrication conditions (1-4). Sintered materials are consisted of silicon nitride grains and glassy grain boundary phase. Crack propagates mainly at grain boundaries, so that mechanical properties depend largely on the size and shape of silicon nitride grains.
One of the most important requirements for the fabrication of high-performance ceramics has been to use α powders (2,5). The development of microstructure is closely related to phase transformation which occurs during sintering. The attempts have been undertaken to accelerate anisotropic grain growth to increase fracture toughness of ceramics (6-8). Gas-pressure sintering has been sometimes applied to facilitate the diffusion at grain boundaries. The microstructures of materials are composed of small number of elongated grains and large number of small grains (9). They are similar to those of whisker-reinforced ceramics, so that they are referred to as in-situ composites or self-reinforced ceramics. The nucleation for the abnormal grain growth occurs during phase transformation (8), so that it was quite difficult to control the microstructure. It has been shown recently that β powder could be sintered to high density (9, 10). The resultant microstructures are uniform in the grain size and shape, because there are large number of nuclei for grain growth. When large β grains are mixed in fine

β powder, in-situ composite microstructure could be developed by selective grain growth of large grains (8). If β powder has wide size distribution, large grains grow preferentially so as to develop in-situ composites (8, 11). It was shown that the size of large grains could be easily controlled in this process. Good reproducibility of the microstructure is shown by high Weibull modulus of sintered materials.
Further investigation on microstructure control should be undertaken in relation to the design of microstructure. If the mechanical properties could be predicted from designed microstructure, we can try further on process control for the optimization of microstructure. Present work intends to summarize the relation between microstructure and mechanical properties for microstructure design. The development of designed in-situ composites is tried by gas-pressure sintering of β powders.

MICROSTRUCTURE DESIGN

Many efforts have been done to improve mechanical properties of ceramics, i.e. strength or fracture toughness. The target microstructures might be divided into two. One is an uniform microstructure and the other is a bimodal microstructure.

Uniform microstructure

The way to make microstructures fine and uniform is a traditional one in oxide structural ceramics. The abnormal grain growth should be inhibited. As silicon nitride grains are basically hexagonal rods, the typical microstructure is shown in Fig. 1. It was obtained by hot-pressing α powder (12). Because of the uniform distribution of grain size, the fracture toughness is low, i.e. about 5 $MPa \cdot m^{1/2}$ or less (marked as ① in Fig. 2). The critical flaw size is minimized by the fine microstructure and high density, so that high strength ceramics are to be fabricated in accordance with Griffith equation,

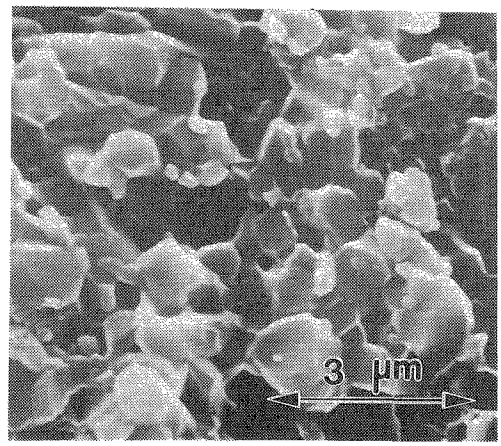

Fig. 1. Microstructure of Griffith materials.

$$\sigma_f = \frac{K_{1C}}{Y\sqrt{c}} \qquad (1)$$

where σ_f, K_{1C}, Y and c is strength, fracture toughness, shape factor and flaw size, respectively. This type of materials will be referred to as Grifith materials (13). The relation between bending strength and flaw size in Griffith materials is shown by Fig. 2. The strength is very sensitive to flaw size. It means that the strength distribution is closely related to the heterogeneity formed during processing. There are a few kinds

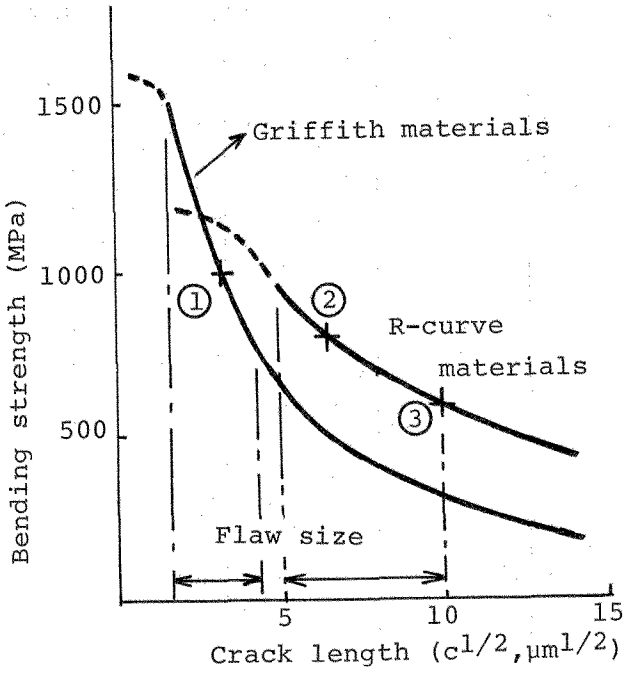

Fig. 2. Effect of R-curve behavior on the relation between strength and crack length.

of strength-limiting flaw, i.e. pores, abnormal grains, foreign particles etc. It is necessary to sinter the powder with fine and narrow size distribution at low temperature to obtain Griffith materials. The fracture toughness of Griffith materials might be represented in Fig. 3 in relation to the crack size.

Bimodal microstructure

It has been shown that the fracture toughness of silicon nitride ceramics increases with the increase in the diameter of large grains which grown in the matrix of small grains (7). A typical microstructure is shown in Fig. 4. The bimodal microstructure formed by gas-pressure sintering at high temperature (6-8). The increase in fracture toughness is suggested to be due to the crack bridging by large grains (7-14). Crack resistance increases with the increase of crack size (Fig. 3). The materials are therefore referred to as R-curve materials. We also refer to these microstructures as in-situ composites, because the microstructures are similar to those of whisker-reinforced ceramics and developed during sintering. The strength dependence of R-curve materials on the crack size is smaller than that in Griffith materials as shown in Fig. 2 (13). It means that the strength distribution of materials could be narrowed in R-curve materials compared with Griffith materials (13). Thus, the fabrication of reliable ceramics is one of the important advantages of R-curve materials.

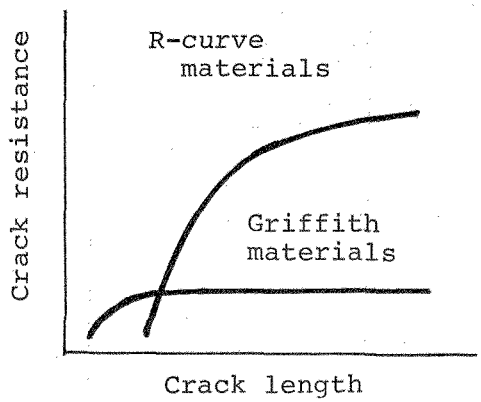

Fig. 3. The crack resistance in relation to crack length.

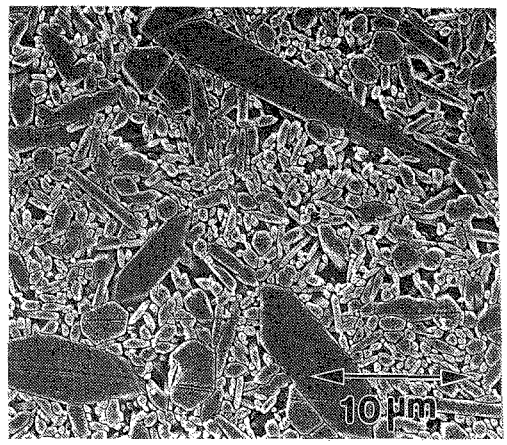

Fig. 4. Microstructure of a R-curve materials.

It can be understood from Fig. 2 that the microstructure for high strength (② in Fig. 2) is different from that for high toughness (③ in Fig. 2) (15). There is a possibility in R-curve materials to develop designed microstructure to control mechanical properties.

FABRICATION OF R-CURVE MATERIALS

The R-curve materials have been fabricated from α powders. The nucleation for large and abnormal grain growth occurs during α to β phase transformation (2). It is not possible to control the amount and size of large grains in materials from α powders. The strength distribution is wide in these materials which results in low Weibull modulud. It was shown recently that size distribution is the main driving force for abnormal grain growth (8). The R-curve materials or in-situ composites could be fabricated from β powders with large size distribution as shown in Fig. 5 which is quite similar to Fig. 4. The materials were gas-pressure sintered at 1900°C. The materials were further heated at 2000°C for 2, 4 or 8 h to grow large grains. The Weibull plots of strength values are shown in Fig. 6. The mechanical properties are also listed in the figure. The average strength is the largest after heating for 2 h. The microstructure is shown in Fig. 7. There are some elongated grains in fine matrix grains. The distribution of large grains is uniform because the nuclei were distributed uniformly and grown preferentially. The good reproducibility of microstructure is shown by high Weibull modulus of 53. The reason for high Weibull modulus is also due to the fact that the fracture originated mostly from one type of flaw, i.e. large grains. The microstructure might be controlled by controlling the number and size of nuclei and the driving force for grain growth.
The sintering for 4 h results in higher fracture toughness of

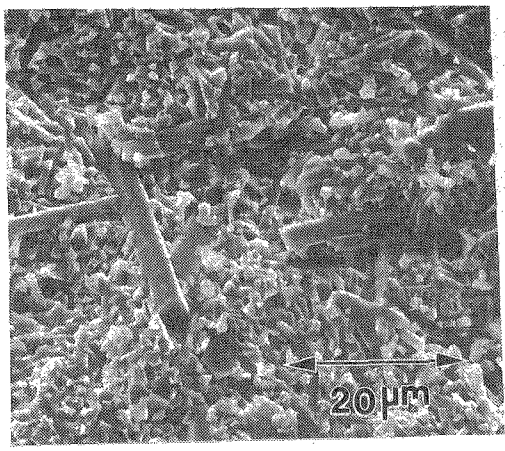

Fig. 5. Microstructure of in-situ composite from β powder.

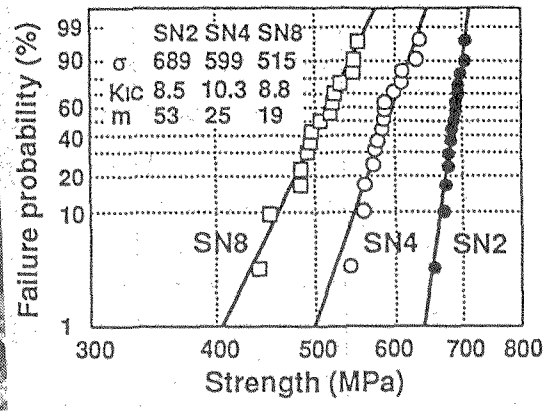

Fig. 6. Weibull plots of strength values.

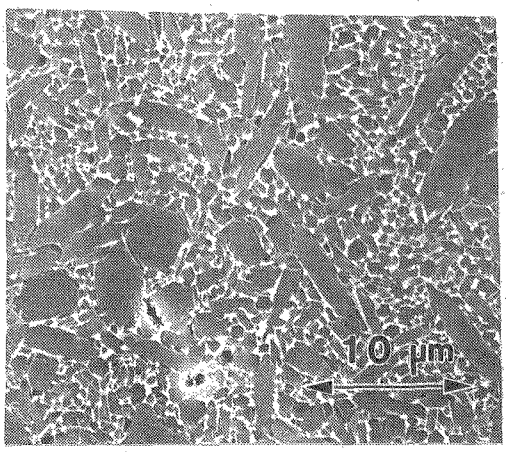

Fig. 7. Microstructure of reliable in-situ composite.

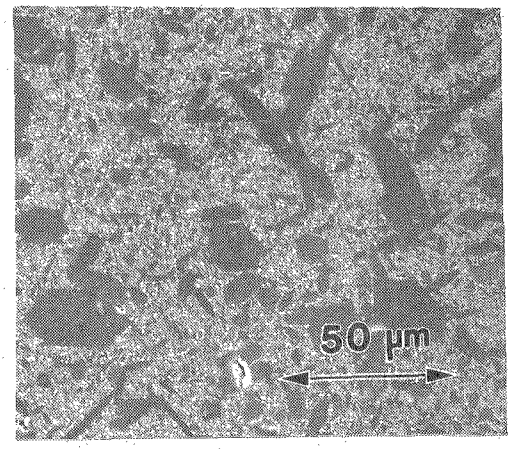

Fig. 8. Microstructure of tough in-situ composites.

10.3 MPa·$m^{1/2}$. It is related to the increase in the average diameter of large grains. The fracture toughness increases by crack bridging with large grains. It is suggested from the theory that fracture toughness increases as proportional to square root of diameter of large grains (14). The strength, however, decreases with the increase of flaw size (Fig. 8). Further heating resulted in the decrease in both strength and fracture toughness. The decrease in fracture toughness might be due to macro-cracking by large residual stresses by thermal expansion mismatch. The Weibull modulus of these materials decreases by wider size distribution of large grains.
It is shown in present work that there is the possibility to fabricate in-situ composites with high strength and high relia-

bility or high fracture toughness by optimizing the size and distribution of large grains. Further investigation is to be done to clarify the effect of processing parameters on the grain growth.

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