High-Temperature Deformation and Fracture Behavior of Al_2O_3 - Y_2O_3 Doped Silicon Nitride

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In order to clarify the high-temperature deformation and fracture behavior of Al_2O_3 - Y_2O_3 doped Si_3N_4 , four-point bending tests were conducted at temperatures from room temperature to 1790K and at strain rates from $1.5 \times 10^{-6} \text{s}^{-1}$ to $3 \times 10^{-2} \text{s}^{-1}$ in a nitrogen gas of $1.013 \times 10^5 \text{Pa}$ (1 atm). As a result, the mechanical equation of state at yield stress was obtained as $\epsilon = A(\sigma_y/E)^m \exp(-Q/RT)$, where $m = 3.9$ in the low temperature range or high strain-rate range, where $m = 1.7$ in the high temperature range or low strain-rate range. The critical strain rate at which m changed shifted to high strain-rate side as temperature rises. It was suggested that plastic deformation of the Si_3N_4 was controlled by grain boundary sliding with cavity formation in the range of $m = 1.7$, and dislocation motion and micro-crack propagation in the range of $m = 3.9$. The activation energy for deformation Q was almost independent of strain rate when $m = 3.9$, while Q tended to increase with decreasing strain rate when $m = 1.7$. The relationship between deformation and fracture in Si_3N_4 was discussed on the basis of m value and fracture mode.

Keywords: silicon nitride, strain rate, stress exponent, grain boundary sliding, cavitation, high-temperature deformation, activation energy for deformation, micro-crack, dislocation

I. Introduction

Ceramics such as Si_3N_4 and SiC are candidate materials for high-temperature structural applications. Although many experiments concerning high-temperature strength of the ceramics have been conducted in the past 10 years⁽¹⁾, plastic deformation behavior of the ceramics has not been paid much attention⁽²⁾ except creep experiments. This is because the amount of plastic deformation in the ceramics is small, comparing with that of elastic deformation until fracture. Even in the case of such ceramics; however, a yielding behavior is frequently observed in the stress strain curves obtained at relatively high temperature⁽³⁾. This indicates that the ceramics fracture after the occurrence of some plastic deformation at the high temperature. Since the plastic deformation behavior of the ceramics is related to the fracture mechanism at the temperature, it is important to clarify the high-temperature deformation behavior of the ceramics.

In this study, high-temperature deformation and fracture behavior of Al_2O_3 - Y_2O_3 doped Si_3N_4 was investigated, with emphasis on the effects of temperature and strain rate of the yield stress.

II. Experimental

Si_3N_4 powders (92.5%- α) were mixed with 5wt.% Al_2O_3 powders and 5wt.% Y_2O_3 powders by ball-milling, and isostatically pressed under about 300MPa for 60s. This green body was sintered at 2120K for 14.4ks in an nitrogen gas of $9.785 \times 10^5 \text{Pa}$ (9.5 atm). The relative density of the sintered body was 97.4 %.

The sintered body was cut into specimens with the size of $3 \times 4 \times 40 \text{mm}^3$ and mechanically polished so as to fit JIS standard method⁽⁴⁾. Load-deflection curves were recorded by the 4-point bending test⁽⁴⁾ in a nitrogen gas of $1.013 \times 10^5 \text{Pa}$ (1 atm) at temperatures from room

temperature to 1790K and at strain rates from $1.5 \times 10^{-6} \text{s}^{-1}$ to $3 \times 10^{-2} \text{s}^{-1}$ (at cross head speeds from 0.005mm/min. to 100mm/min.). The maximum fiber stress was used as stress, and strain was corrected by subtracting the machine stiffness⁽⁵⁾. Young's modulus was estimated by the static method⁽⁴⁾ and the yield stress was obtained by the proportional limit of stress-strain curve.

Microstructure of the samples before and after bending tests was observed with transmission electron microscope (TEM), scanning electron microscope (SEM) and optical microscope. JEOL 4000FX operating at 400kV and Hitachi S-800 operating at 20kV were used for TEM and SEM observation, respectively. The tensile surface near the fracture surface was cut out for TEM observation of the samples after bending tests. The samples for TEM observation were thinned by ion beam sputtering at a voltage of 5kV using a JEOL JIT-100.

III. Results and Discussion

1. Microstructure

Fig.1 shows the SEM micrograph of a specimen treated by plasma-etching⁽⁶⁾, indicating that white contrast corresponds to grain boundary phase. The grain size distribution of the sample was from $0.1 \mu\text{m}$ - $3 \mu\text{m}$.

Fig.2 shows (a) a high resolution electron micrograph of a grain boundary in the Si_3N_4 . It is noted that the thickness of the boundary phase between two crystals is approximately 2 nm. Micro-diffraction pattern and energy dispersive spectroscopy (EDS) spectrum taken from a triple junction phase denoted by T indicated that the intergranular phase is amorphous which was composed of Al, Y, O, Si and Ca.

2. High-temperature deformation

Fig.3 shows the stress-strain curves deformed at various strain rates from $1.5 \times 10^{-6} \text{s}^{-1}$ to $3 \times 10^{-2} \text{s}^{-1}$ at 1660K. It is noted that the yield stress increases with increasing strain-

Fig.1 SEM micrograph of the $\text{Y}_2\text{O}_3\text{-Al}_2\text{O}_3$ doped Si_3N_4 treated by plasma-etching, indicating that white contrast corresponds to grain boundary phase.

Fig.2 High resolution electron micrograph of a grain boundary in the Si_3N_4 , indicating the presence of second phase along the boundary. Triple junction phase for micro-diffraction and EDS analysis is indicated by T.

Fig.3 Stress-strain curves for the Si_3N_4 bent at various strain rates at 1660K. Note that yield stress indicated by arrows depends on strain rate.

rate as shown by the arrows. The specimen bent at a high strain rate of $1.5 \times 10^{-2} \text{ s}^{-1}$ does not show yielding behavior to be fractured in a brittle manner even at the high temperature of 1660K. This means that the yield stress is higher than fracture stress at this strain rate.

Temperature dependence of fracture and yield stress at a strain rate of $1.5 \times 10^{-4} \text{ s}^{-1}$ is shown in fig.4. Fracture strength was about 730MPa until 1170K, but decreases rapidly above 1170K as shown by white circles, while yield stress of plastic deformation was seen only above 1520K as shown by black circles. This suggests that fracture strength becomes higher than yield stress at above approximately 1520K at this strain rate.

Fig.4 Temperature dependence of fracture (white circles) and yield stress (black circles) at the strain rate of $1.5 \times 10^{-4} \text{ s}^{-1}$.

Fig.5 $\text{Log}(\sigma_y/E)$ vs. $\text{Log} \dot{\epsilon}$ at various temperature, indicating that the dependence is expressed by two kinds of straight lines.

Fig.5 shows the double-logarithmic plot of yield stress divided by Young's modulus σ_y/E against strain rate $\dot{\epsilon}$ at various temperatures. Here, yield stress is normalized by Young's modulus measured by static method at each

temperature⁽⁷⁾. It is found that the relationship between σ_y/E and $\dot{\epsilon}$ can be expressed by two kinds of straight lines. That is, $\dot{\epsilon}$ is proportional to $(\sigma_y/E)^m$, where m is the value called stress exponent. Since the reciprocal of the slope of straight lines in fig.5 corresponds to the stress exponent m , m is estimated to be 3.9 in the high strain rate and low temperature range, and 1.7 in the low strain rate and high temperature range. This indicates that m changes at a strain rate as shown by dotted line. This critical strain rate shifts to high strain rate side with increasing temperature.

Fig.6 Log(σ_y/E) vs. $1/T$ at two kinds of strain rates, indicating that the dependence is described by two kinds of straight lines.

Fig.6 shows the temperature dependence of yield stress divided by Young's modulus at different strain rates. This relation is also described by two kinds of straight lines. An apparent activation energy for deformation Q can be estimated from the slope in this figure. The slope seems to change at a temperature; however, this change is in fact

mainly affected by that of stress exponent m , that is to say, m equals 3.9 where the slope is low, and 1.7 where the slope is high, respectively, as shown in fig. 6.

From the results of figures 5 and 6, the mechanical equation of state at yielding for the Si_3N_4 can be expressed as $\dot{\epsilon} = A(\sigma_y/E)^m \exp(-Q/RT)$, where A is a constant, m : the stress exponent of strain rate, Q : an apparent activation energy for deformation, R : gas constant and T : absolute temperature. According to this equation, where $\dot{\epsilon}$ is equal to $1.5 \times 10^{-4} \text{ s}^{-1}$, $Q = 269 \text{ kJ/mol}$ in the range of $m = 1.7$ and $Q = 253 \text{ kJ/mol}$ in the range m equals 3.9, and when $\dot{\epsilon} = 1.5 \times 10^{-5} \text{ s}^{-1}$, $Q = 303 \text{ kJ/mol}$ in the range of $m = 1.7$ and $Q = 261 \text{ kJ/mol}$ in the range of $m = 3.9$. This indicates that the Q value is almost independent of strain rate in the range of $m = 3.9$ and tends to increase with decreasing strain rate in the range of $m = 1.7$. The Q values obtained by common creep tests have been reported to be from 360 to 850 kJ/mol⁽⁸⁾. These values are larger than those obtained in this study, however, the strain rate in this study is higher than that of creep tests. The fact that Q tends to increase with decreasing strain rate in the range of $m = 1.7$ is likely to suggest that Q increases up to the values reported in the creep tests. In this case, strain rate is considered to affect an apparent activation energy for deformation.

It is reported that the stress exponent m estimated by creep test is from 1 to 2.3 for Si_3N_4 ⁽⁹⁾⁽¹⁰⁾. In the case of diffusion creep and viscous flow of grain boundary phase, m is considered to be 1, and in the case of grain boundary sliding with cavity formation, m is considered to be 2⁽¹¹⁾. The behavior in the range of $m = 1.7$ in this study may correspond to the range in which creep test is conducted. On the other hand, when dislocation motion affects deformation mechanism, stress exponent m is reported to be larger than 3⁽¹¹⁾. In the range that $m = 3.9$ in this study, dislocation may play some role for deformation. These are discussed in detail at the next section.

3. Microstructure and deformation mechanism

Fig. 7 shows the back scattered electron images of fracture surface in the specimens bent at (a) 1290K, (b) 1570K and (c) 1790K at a strain rate of $1.5 \times 10^{-4} \text{ s}^{-1}$. As seen in the

Fig. 7. Back scattered electron images of the fracture surface in the Si_3N_4 bent at (a) 1290K, (b) 1570K and (c) 1790K at the strain rate of $1.5 \times 10^{-4} \text{ s}^{-1}$.

series of micrographs, transgranular fracture was always observed in the specimens bent at low temperature as shown in (a). While almost the perfect intergranular fracture was observed in the specimens deformed in the range $m=1.7$ as shown in (c). In this case, the fracture surface was macroscopically uneven. In the range of $m=3.9$, the fracture surface was fairly flat, and partial transgranular fracture was observed as shown in (b).

Fig.8 A bright-field TEM micrograph of the specimen bent at the strain rate of $1.5 \times 10^{-4} \text{ s}^{-1}$ at 1790K, showing the occurrence of grain boundary cavitation.

Fig. 8 shows typical TEM micrographs in a tensile surface of the specimen deformed in the range m equals 1.7. Grain boundary cavitation was frequently observed in this range as shown in (a). According to the results obtained by creep tests (2), when creep is due to grain boundary sliding with cavity formation, the stress exponent is reported to be approximately 2 which is near value to 1.7 in this study. Therefore, the plastic deformation in the range m equals 1.7 is considered to be controlled by grain boundary sliding with cavity formation. This explanation is consistent with the fact that intergranular fracture is predominant in this region. Since cavities are formed at fairly wide region to link each other, the fracture surface is likely to become uneven.

Fig. 9 shows typical TEM micrographs in the specimen deformed in the range m equals 3.9, indicating that micro-crack through a grain boundary (a) and dislocation inside a grain (b). As mentioned at previous section, when dislocation motion affects the creep mechanism, the stress exponent is reported to be larger than 3. If plastic deformation in this range is controlled by dislocation motion, the stress exponent 3.9 might be reasonable; however, yielding point appeared in the stress-strain curve is not believed to be due to only pure plastic deformation because the density of dislocation is not so high. An alternative explanation for yielding point is that yielding behavior may attribute micro-crack propagation which is induced by stress concentration of dislocation accumulation at grain boundaries. In this case, the micro-crack is thought to be propagated by cutting Si_3N_4 grains; however, crack deflection may occur through the grain boundary in the case

Fig.9 TEM micrographs in the specimen bent at the strain rate of $1.5 \times 10^{-4} \text{ s}^{-1}$ at 1570K, indicating that micro-crack through a grain boundary (a) and dislocation inside a grain (b).

of crystal orientation where a grain is hard to cut. This agrees well with the fact that both inter and transgranular fracture occurs in this region. As micro-crack propagates across the specimen with keeping minimum crack pass, the fracture surface in this region may become flat.

As mentioned above, in the case of sintered Si_3N_4 , fracture mode was found to be related to stress exponent at high-temperature.

IV. Conclusions

High-temperature deformation and fracture behavior of $\text{Al}_2\text{O}_3\text{-Y}_2\text{O}_3$ doped Si_3N_4 was investigated by four-point bending test systematically, and their microstructure was characterized by electron microscopy. The results obtained are summarized as follows.

1. The mechanical equation of state at yielding is described as $\dot{\epsilon} = A(\sigma_y/E)^m \exp(-Q/RT)$.
2. The stress exponent m is 3.9 in the high strain rate and low temperature range, and 1.7 in the low strain rate and high temperature range.
3. An apparent activation energy for deformation Q in the range m equals 3.9 is about 260kJ/mol, although Q tends to increase with decreasing strain rate in the range m equals 1.7.
4. Plastic deformation in the range m equals 1.7 is considered to be controlled by grain boundary sliding with cavity formation. Plastic deformation in the range m equals 3.9 is considered to be controlled by dislocation motion and micro-crack propagation.
5. Fracture mode is related to stress exponent at high-temperature.

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