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Crack-wake Shielding and Small-scale Crack-tip Yielding as Potential Mechanisms for Improving the Delayed-failure Resistance of Silicon Nitride at Elevated Temperatures

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Abstract: Requirements and microstructural design criteria for employing silicon nitride in long-term structural applications at elevated temperatures are discussed according to fracture mechanics concepts. Three least engineering parameters are considered: lifetime, flaw-tolerance and deformation under stress. "Ductile" materials are found by exploitation of small-scale crack-tip yielding which arises from the softening of their grain-boundary phase. These materials, however, are likely to exhibit poor deformation resistance. Materials with a "strong" grain boundary generally show a superior deformation behaviour but are liable to brittle fracture and static-fatigue strength degradation unless shielding mechanisms in the crack-wake be operative. The present analysis evaluates and compares these two classes of materials and the respective approaches commonly followed for their densification. © 1996 Elsevier Science Limited and Techna S.r.l.

1 INTRODUCTION

Traditional design in the field of structural metallic materials usually relates to the elastic range of deformation of "ductile" materials through the assessment of a safety-factored yield stress. This design philosophy has been as often as not coupled with boundary calculations by linear elastic fracture mechanics in order to permit the use of high strength materials whose toughness/yield-stress ratios are much lower than those of the traditional ductile solids. In principle, a similar approach can also be suitable and has been aimed in ceramics for structural applications. Thus, the "need" of a yield stress for design has been the key concept for developing structural ceramics as, for example, the Si_3N_4.

The fundamental requirements for employing structural ceramics at elevated temperatures have been recently reviewed by Raj.[1] In this article, we will restrict our treatment to Si_3N_4 ceramics and examine two basic approaches for achieving a "yield" behaviour, namely an acceptable design reliability, at elevated temperatures. One approach, which has also been the most commonly pursued. so far, is related to the obtainment of the small-scale crack-tip yielding effect arising from the softening of the grain-boundary phase. Another approach, only recently explored, deals with a shielding effect by bridging forces in the wake of brittle cracks. Previous studies[2-6] have been focussed on these subjects and indicated that the beneficial effects by crack-tip yielding and crack-wake shielding can be individually obtained but are hard to be profitably superimposed.

2 LIFETIME OF Si_3N_4 CERAMICS AT AROUND 1400°C

2.1 "Ductile" materials by small-scale crack-tip yielding

Development of Si_3N_4 for structural applications such that high strength must be retained at high temperature for long-term operation has been the object of considerable effort. Basically, at the temperatures and tensile stresses hypothesized for a profitable employment (i.e. 1400–1600°C, < 500 MPa), the Si_3N_4 grains resist deformation and have no dislocation mobility. Thus, an inherent material plasticity can only be achievable through the softening of a grain-boundary phase. This phenomenon has been found to give rise to the blunting of pre-existing flaws and, hence, to provide a yield stress. So far, however, materials doped with softer phases (for example, oxides to form, during sintering, eutectics with SiO_2 at the grain boundaries) for achieving such ductility, have often been lacking deformation resistance and are liable to severe intergranular cavitation phenomena. Accordingly, the limited lifetime and the poor adaptation to strict dimensional tolerances have constituted serious problems for their actual employment. In order to overcome these problems, research efforts has been directed to the crystallization of the grain-boundary phase. Figure 1 shows the results of lifetime tests at 1400°C for various selected Si_3N_4 ceramics sintered with oxide additives. Particularly, materials containing only small volume fractions of amorphous grain-boundary phase (densified by hot-isostatic pressing (HIP)[3,5]) are compared to an optimized (pressureless) sintered Si_3N_4 material containing large amount of crystallized secondary phase (Kyocera SN251[7]). It is noteworthy that, despite the large fraction of grain-boundary phase and due to their crystallization process, the latter material can be loaded at high stress levels and show remarkable lifetimes.

2.2 "Brittle" materials with shielding forces in the crack-wake

Si_3N_4 materials containing only pure SiO_2 at the grain boundaries (densified by HIP) generally show a high resistance to the formation of cavities at 1400°C but also a negligible crack blunting effect. Thus, they behave in "brittle' fashion even at elevated temperatures.[3–6] Their lifetime at high temperature is limited mainly by the subcritical-crack-growth (SCG) of inherent flaws or damages suffered during operation. Figure 2 shows data for selected Si_3N_4 materials with an SiO_2 grain-boundary. The most important features dealing with this kind of material can be summarized as follows: (1) the log–log plot of static lifetime vs stress is generally found to be a straight line as opposed to the sigmoidal curves in Fig. 1. This means that the high-temperature crack-growth law can be phenomenologically expressed by a power-law equation with an exponent n representing the SCG rate of the material (this point has been treated in detail elsewhere[4,8,9]); (2) the addition of small fractions of pure SiO_2 does not degrade significantly the lifetime of the material at 1400°C (as compared with the materials displayed in Fig. 1 which are doped with similar volume fractions of oxides other than SiO_2 to form eutectic phases

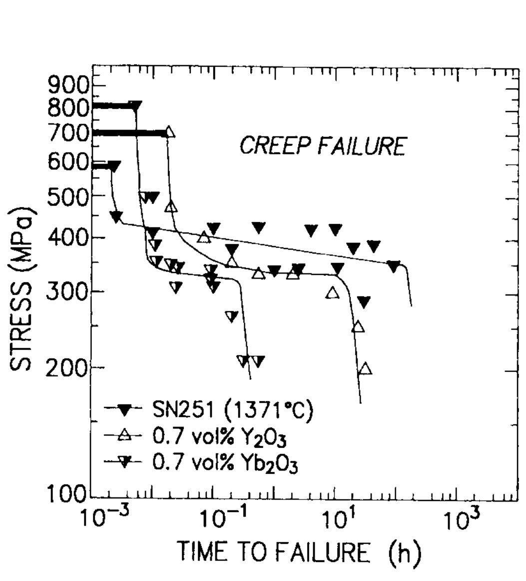

Fig. 1. Results of lifetime test (1400°C, in air) for materials containing oxide additives. Data for the SN251 material are from Ref. 7.

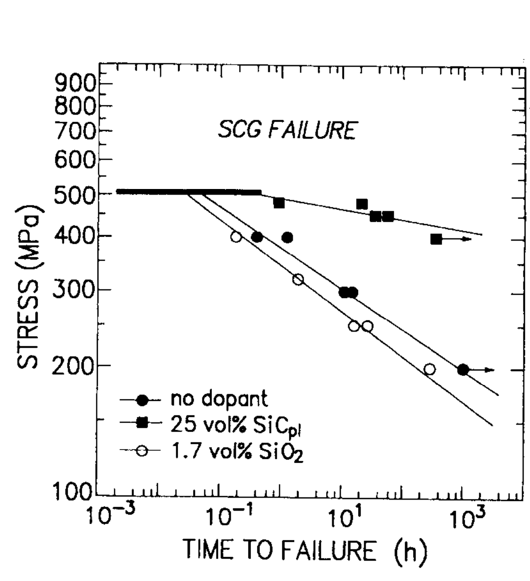

Fig. 2. Results of lifetime test (1400°C, in air) for materials with pure SiO_2 at the grain boundary. Arrows locate the specimens which did not fracture at the time of the test interruption.

at the grain boundaries); (3) a fibrous microstructure (provided, for example, by adding an appropriate fraction of SiC platelets (Grade M, C-axis operating crack-wake bridges in the presence of a highly viscous/pure SiO_2 interface, can markedly delay the propagation of static-fatigue cracks and even arrest them up to high stress levels. It should be mentioned that, conceptually, the micromechanisms leading to shielding forces in the crack-wake, if operative in a small process-zone (i.e. in a short crack-length extent) behind the crack-tip, can provide a pseudo yield stress for the material. This phenomenon, which appears to be particularly beneficial in obstructing the propagation of high-temperature static-fatigue cracks (Fig. 2), has also been found to provide some improvement in the fast-fracture resistance of the material.[2]

3 FLAW-TOLERANCE OF Si_3N_4 CERAMICS AT 1400°C

In order to obtain information on the flaw tolerance at high temperature, strength measurements were carried out on bars pre-cracked with semi-elliptical flaws of increasing depth. The results obtained at 1400°C are plotted in Fig. 3 for the same materials displayed in Figs 1 and 2. All the materials were loaded at a strain rate of $1.5\times10^{-7}s^{-1}$ to monitor their SCG behaviour with the exception of the SN251 (i.e. the one containing the crystallized grain-boundary phase). In this latter case, it was not possible to load the specimen at such a low strain-rate since the material was liable to severe deformation at very low stress levels and failed by a continuous flow. For discussing also the flaw-tolerance of this material, data at the higher strain rate of 1.5×10^{-5} s^{-1}

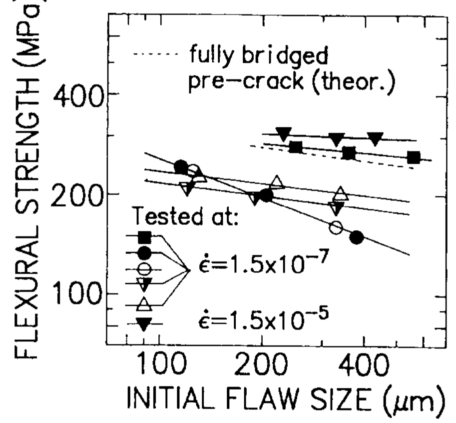

Fig. 3. Strength (at 1400°C) as a function of the size of initial pre-crack for various materials. The strain rate $\dot{\epsilon}$ is in s^{-1}.

are shown. It should be noticed that the SN251 material did not fail from the pre-cracks, whatever their size in the investigated range shown in Fig. 3. Further, this material, undergoing mainly creep failure independently of the inherent flaw size and distribution, showed yield and fracture stress at almost constant levels.

As a general trend, the materials containing dopant other than pure SiO_2 at the grain boundary showed better flaw tolerance as a consequence of the relief of the local stress field at the crack-tip due to plasticity. However, as an exception among Si_3N_4 with interfaces constituted by pure glassy SiO_2, almost no strength degradation with increasing flaw size was found in the composite reinforced by SiC platelets. Its high-temperature flaw-tolerance was similar to that of the SN251 even by testing at two orders of magnitude lower strain rate. The micromechanism behind such a behaviour is the same as that producing the lifetime elongation shown in Fig. 2 and is related to the shielding forces operated by the SiC platelets in the crack-wake.

The dependence of the material strength on the initial flaw-size in presence of crack-wake shielding and under SCG regime can be theoretically predicted by a fracture mechanics approach. This method, based on an algorithm originally proposed by Lawn,[10] has been developed and described in detail in a previous report.[6] When the fracture stress σ_{fm} of the matrix, its SCG exponent n and the frontal factor R of the composite R-curve equation are known, the fracture stress σ_{fc} under loading at a constant stressing rate (Fig. 3), can be calculated according to the following system of equations:

$$\frac{1}{\sigma_{fc}}\int_{a_{ic}}^{a_{fc}}\frac{da}{[\sigma_{fc}Ya - Ra]^n} = \frac{1}{Y^n\sigma_{fm}^{n+1}}\int_{a_{im}}^{a_{fm}}\frac{da}{a^{n/2}}, \qquad (1)$$

$$\sigma_c = \left[\frac{1}{n+1}\right]^{\frac{1}{n}}\sigma_{fc}, \qquad (2)$$

where a_i is the initial flaw-size (abscissa in the plot of Fig. 3) and a_f is the crack length at the onset stress for fracture which can be directly measured on the fracture surface or calculated from the value of fast fracture strength through the Irwin's similarity relation.[2,6] The subscripts m and c locate the parameters dealing with the monolithic and composite material, respectively. Y is a geometrical parameter related to the pre-crack geometry and the stresses σ_{fm} and σ_{fc} are the static stresses, respectively, for matrix and composite, equivalent to the fracture stresses σ_m and σ_c applied during

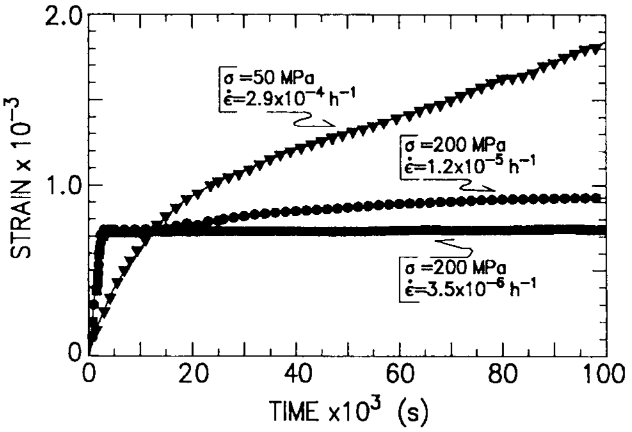

Fig. 4. Creep curves for various materials at 1400°C.

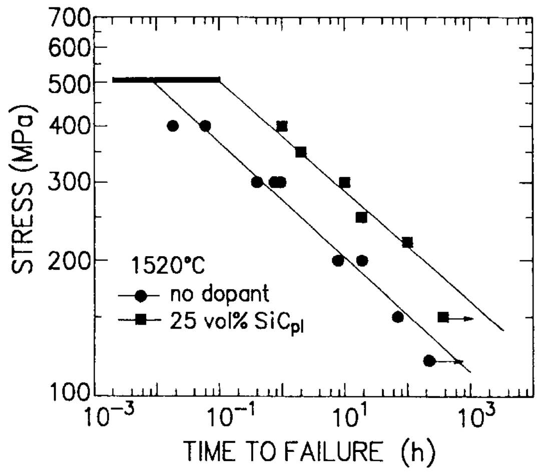

Fig. 5. Results of lifetime test (1520°C, in air) for materials with pure SiO_2 at the grain boundary. Arrows have the same meaning as in Fig. 2.

dynamic loading at a constant strain rate.[8] After substituting in eqn (1) the values[9,11,6] $n=20.7$ and $R=4.79$ MPa·m$^{1/2}$/mm, and calculating σ_m from the experimental strength values σ_{fm} in Fig. 3 (by means of eqn (2) with the appropriate subscripts), eqn (1) can be solved numerically to give σ_c and thus σ_{fc} (via eqn (2)). Note that, according to the left-hand integrand in eqn (1), the case examined here corresponds to a fully bridged indentation pre-crack. The theoretical plot of the strength vs the initial flaw-size (as calculated according to eqns (1) and (2) is shown in Fig. 3 and is found to fit well the experimental data, a result which explains the important role of the crack-wake shielding mechanisms on the high-temperature reliability of Si_3N_4 and provides an important criterion for microstructural design.

4 DEFORMATION BEHAVIOUR OF Si_3N_4 CERAMICS AT 1400°C

The deformation behaviour of the SN251 sample, already mentioned with regard to the strength behaviour at low strain rates on pre-cracked bars (Fig. 3), was investigated in detail by creep tests. Figure 4 shows the strain vs the loading time under a nominal (bending) stress of 50 MPa at 1400°C. As a result of the large primary-creep strains, this low stress level represents the maximum value for which the steady-state creep rate could be reasonably measured in bending. Despite the crystallization of the grain-boundary phase, a poor deformation resistance was peculiar to this material, suggesting serious difficulties in any application requiring adherence to strict tolerances.

Materials with the SiO_2 intergranular phase showed very small primary-creep deformation and creep rate (see Fig. 4) behaving virtually as Griffith materials at elevated temperature. The strain rate of the composite containing SiC platelets was found to be exceptionally low, also suggesting the important role of the brittle SiC phase in improving the intrinsic creep resistance of Si_3N_4.

5 LIMIT AT AROUND 1500°C FOR Si_3N_4 CERAMICS WITH GRAIN BOUNDARIES

Increasing the testing temperature up to 1520°C, the degradation phenomena related to the grain boundaries becomes extremely severe and the mechanical responses of different materials more markedly differentiated. The Si_3N_4 with glassy-SiO_2 at the grain boundary still show linear behaviour but also a noticeable drop down in the lifetime under constant stress (Fig. 5). The inherent properties of the SiO_2 intergranular film as, for example, its viscosity or volatility in presence of oxygen,[1] dominate the material behaviour at 1520°C and also the addition of SiC platelets becomes much less effective in elongating the material static lifetime. Note in Fig. 5 that the slopes of the lifetime plot, which is related directly to the SCG exponent n, become very close for the monolithic and composite materials, suggesting that the crack-wake shielding mechanism can be fully exploited only when operating in highly SCG resistant matrix phases.[6]

The Si_3N_4 with the crystallized grain-boundary phase loses dimensional stability and behaves as a viscoelastic solid in which flow and fracture are combined depending upon the amount of plastic deformation. Figure 6 shows the crack-growth behaviour of this material. The plot was obtained according to a procedure originally proposed by Ohji:[12] a bend bar was deeply notched in the centre

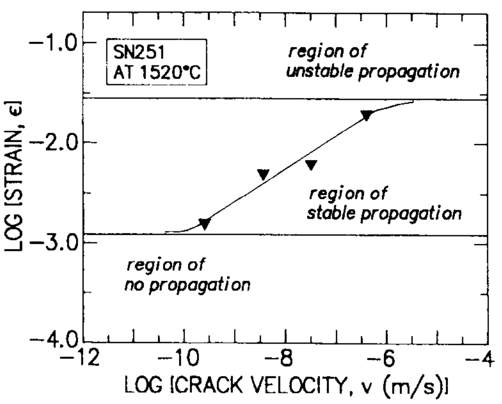

Fig. 6. Crack-growth behaviour for the SN251 material at 1520°C, in air.

and loaded in four-point geometry by dead weight monitoring the apparent strain of the material (i.e. the creep compliance plus the compliance due to the notch) via a computer as in a usual creep test. Then, at certain strain values, the load was increased at very high speed to achieve instantaneous fracture. After cooling down the specimen, the portion of crack length extended under constant load was easily recognized on the fracture surface due to the different fracture mode and measured by an optical microscope. The crack velocity was simply obtained by the ratio of the measured crack length over the loading time. During crack growth in the SN251 Si_3N_4 material at 1520°C, the crack was extended by plastic separation of the ductile grain boundary and the macroscopic mechanical response of the material was that of a linearly viscoelastic material.[13]

6 CONCLUSION

In this article, we have examined two different approaches for achieving an acceptable structural reliability, namely a high flaw tolerance and a high value of "yield stress", in Si_3N_4 materials at around 1400°C. An appreciable crack blunting effect via the softening of the grain-boundary phase (found only in some optimized material containing crystalline silicates (i.e. SN251)[7]), could only be obtained at the expense of the deformation behaviour of the material. A more promising approach was instead recognized in exploiting crack-wake shielding forces operated by a brittle SiC phase (in the shape of platelets) in presence of a highly viscous glassy-SiO, grain-boundary film. By this latter approach, increased flaw tolerance and good deformation resistance at 1400°C could both be attained. However, a limit for Si_3N_4 with an oxide film at the grain boundaries seems to reside at around 1500°C because of the severe SCG degradation. This important circumstance calls for new approaches in order to densify Si_3N_4 materials in which the grain-boundary film has been eliminated.

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