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Dynamic Fatigue of Alumina Ceramics in Water-Containing Environment

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Abstract: Alumina ceramics have found wide application in water-containing media being subjected to delayed failure resulting from the stress-enhanced chemical reactions between environment and ceramics. Effect of composition of both the environment and ceramics on delayed fracture of various alumina-base ceramics is investigated. A dynamic fatigue technique was utilised with special emphasis on proper choice of the experimental conditions for testing procedure. High-alumina ceramic material is revealed to be more resistant to subcritical crack growth than glass-bonded materials. The behaviour of glass-bonded ceramics is dependent on the composition of glassy grain-boundary phase. The water content in environment affects slightly the crack velocity exponent and influences the mean crack-growth velocity. The results are discussed in terms of the chemical reaction rate. © 1998 Elsevier Science Limited and Techna S.r.l. All rights reserved

1 INTRODUCTION

Alumina ceramics are good candidates for application in water-containing media, e.g. in seal-ring devices, except for their low resistance to subcritical crack growth. This phenomenon is believed to result primarily from a stress-enhanced chemical reaction between a chemical environment and the bonds in ceramics.[1–4] As a consequence, strength of alumina-base materials is dependent on time, or on stressing rate. This effect must be taken into account when designing with ceramic parts.[5]

Generally, there are three regions of the kinetic crack propagation diagram, each of the three regions results from different mechanisms of crack growth (Fig. 1). Region I of crack-growth behaviour is empirically described by the relation between crack-growth velocity, v, and the applied stress intensity factor, K, as follows:

$$v = AK^n \qquad (1)$$

where A is a constant and n is the crack velocity exponent. The crack-growth rate is reaction-rate limited in this region. Region I is the main contributor to the lifetimes of ceramic material. Obviously, the parameters of the kinetic eqn (1) have to be affected by composition of both the ceramic material and environment. In this context, the present work was aimed at investigation of delayed failure of various alumina-base ceramics to reveal the effects of environment and grain-boundary phases on the crack-velocity exponent.

2 EXPERIMENTAL

Experiments were performed with the ceramic materials prepared by cold uniaxial pressing followed by sintering. Three series of the materials were studied:

1. Dense 99.5 wt% Al_2O_3 ceramics with 0.5 wt% MgO as sintering additive.
2. The 88 vol% Al_2O_3 ceramics with 12 vol% grain-boundary glassy phase, the latter was in the system Me'_2O–$Me''O$–B_2O_3–SiO_2, where Me' is Na and K, Me'' is Mg, Ca and Ba. After sintering the density of the material was about the theoretical one, as measured by the water-displacement method. There were elongated alumina grains having length of about

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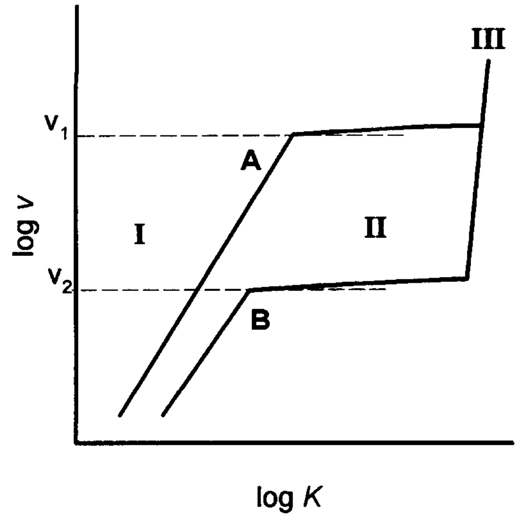

Fig. 1. Kinetic v-K diagram: I, II and III—the stages of crack-growth law; A—in water environment, B—in humid air; v_1 and v_2 are the critical crack-growth velocities.

15–40 µm and thickness of about 7–15 µm in the microstructure. These crystals are surrounded by glassy-phase layers, the latter being about 2 µm thickness.

3. The full-dense 85–89 vol% Al_2O_3 ceramics bonded with grain-boundary phase in the system Y_2O_3-Al_2O_3-SiO_2. After sintering, the length of alumina grains was *ca.* 5–35 µm.

To study the delayed failure, a dynamic fatigue test method was chosen, where strength is measured as a function of deformation rate.[67] This method is becoming an increasingly popular test for measuring the crack velocity exponent for ceramic materials. Data from the dynamic fatigue test are fitted to the equation:

$$\log\sigma = C + [1/(1+n)]\log\varepsilon' \qquad (2)$$

where σ is the measured strength; ε' is the straining rate; C is a constant. The value of n is determined from eqn (2) by the least-squares fit of the data.

The specimens of $7\times8\times45\,mm^3$ size were loaded in a stiff 3-point loading adjustment at a span 32 mm in ambient air atmosphere at relative humidity RH = 59–62% and a temperature 20–22°C, or in water. In the latter case, the specimens were loaded immediately after immersing them into the water. A UTS-100 screw-drived machine was utilised. The cross-head speed of testing machine was in the range from 0.01 to 5 mm.mn^{-1}.

To estimate the crack-velocity exponent value, the proper choice of the strain rates range is of great importance. Only the strain rates should be used which are lower than the certain critical stain rate corresponding to the transition from stage I to stage II of the subcritical crack propagation diagram (Fig. 1). This critical straining rate can be determined using a point of the log σ—log ε' plot where the slope of the plot is changing from the linear dependence of log σ on log ε' to about constant values of log σ with an increase in straining rate. According to Fig. 1, the critical strain rate is dependent on the environment while the crack-growth exponent value can be the same in environments with different water content, for example.

3 RESULTS AND DISCUSSION

Figures 2 and 3 show the dynamic fatigue data for the specimens of A1 series tested in humid air and in water, respectively. Obviously, the strength increases with an increase in strain rate. A fit of the experimental data to eqn (2) gives the values of n equal to 60 and 54 for the specimens tested in air and in water, respectively. The squared value of the correlation coefficient r was estimated to be in the range 0.41–0.50 for all plots, including these for A2 and A3 series. To compare, n values of 35–68 have been reported for alumina in an aqueous environment,[6] 67 for alumina—12 wt% zirconia ceramics in air,[7] and 80 for pure pore-free hydroxyapatite ceramics in humid air.[8]

Shown in Fig. 4 are the data for the A2 series tested in water. Estimated value of n was about 39. The data for the A3 series are given in Figs 5 and 6. It can be seen that there is two-stage dependence of the strength on straining rate for the A3 series tested in humid air (Fig. 5). Evaluated for low-straining rates region, the crack velocity exponent value was equal to 36 in humid air. In water environment, the value of n was evaluated to be about 27.

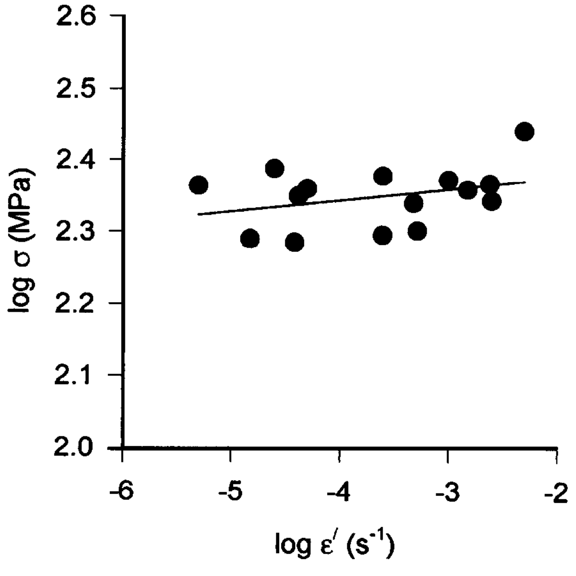

Fig. 2. Strength vs straining rate plot for the A1 series tested in humid air.

Therefore, the water content in environment decreases slightly the crack-velocity exponent on stage I of the kinetic crack propagation diagram, and can affect the mean subcritical crack velocity, just because of its dependence on the position of the point of transition from stage I to stage II on the crack propagation diagram (Fig. 1). This can be concluded, at least, by comparing the data for the A3 series tested in air and in water, although further experiments are needed to quantify this phenomenon.

The glassy phase increases significantly the sensitivity of alumina ceramics to subcritical crack growth making ceramics less resistant to fatiguing. It is known that the silica-containing phase can interact actively with water, and stress concentration at the crack tip enhances this effect.[9] The incoming water molecule interacts with the Si–O–Si crack-tip bond in three stages.[10] A model of this interaction has been proposed in Ref. 10. However, the presence of cations other than silicon must complicate the process. Particularly, the composition of the grain-boundary phase influences the pH value of environment at the crack tip. According to Ref. 1, the components of the ceramics reacting with water at the crack tip probably control the pH of the solution at the crack tip. This reaction may create an acidic or basic environment, e.g. ion-exchange between the alkali ions in the grain-boundary phase of ceramics and the hydrogen ions in the water creates a highly basic solution at the crack tip. The glasses containing highly basic

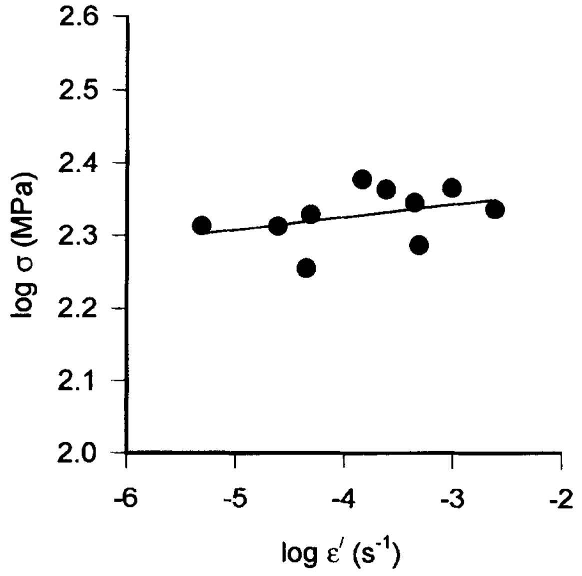

Fig. 3. Strength vs straining rate plot for the A1 series tested in water.

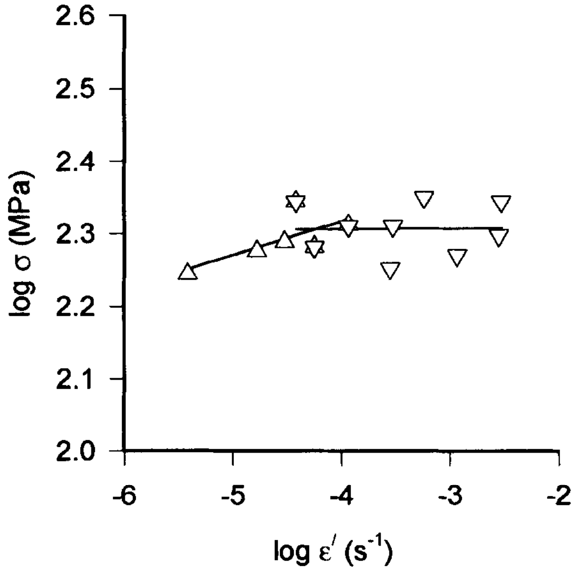

Fig. 5. Strength vs straining rate plot for the A3 series tested in humid air.

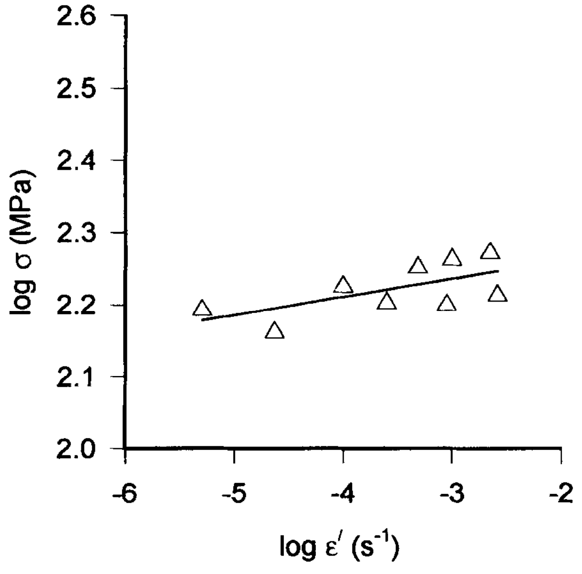

Fig. 4. Strength vs straining rate plot for the A2 series tested in water.

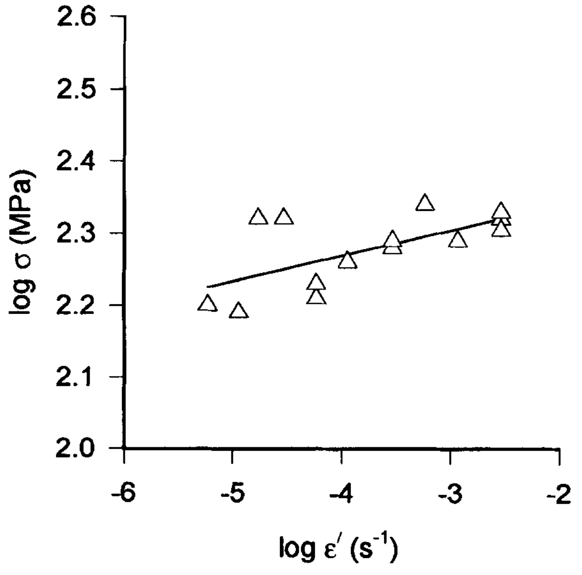

Fig. 6. Strength vs straining rate plot for the A3 series tested in water.

components such as alkali ions were shown to have a crack-velocity exponent that was substantially higher than glasses such as silica that contain no basic constituents.[1] Taking this into account, it is becoming obvious why the alkali-glass containing A2 series of the specimens are more resistant to the delayed failure in the water than silica-yttria-alumina bonded ceramics of A3 series. However, comparing the data for the 99% alumina ceramics and for the glass-bonded aluminas, it can be pointed out that the content of the glassy phase is probably the main factor influencing the resistance of alumina ceramics to delayed failure in water-containing environments.

To discuss the effect of water content on the crack-velocity exponent, speculations on the base of the chemical reactions rate theory can be utilised. The crack-growth rate at region I of the crack propagation diagram is assumed to be proportional to the rate of the reaction of ceramics with water in the environment. If the reaction is assumed to be first order, so the crack-growth velocity can be expressed as follows:[3]

$$v = v_0 a_s \exp(\mu_{s0} + \mu_g - \mu_0^*)/RT \qquad (3)$$

where v_0 incorporates the frequency factor, the activity coefficient, and geometric factors relating the number of bonds broken; as is the activity of the water; the chemical potentials μ_{s0}, μ_g and $\mu{*}_0$ refer to the water, the reactive species in the grain-boundary phase and the activated complex, respectively; R is gas constant, and T is temperature. The chemical potentials are not all equivalent with regard to water-enhanced crack growth. The chemical potentials of both the glass-phase and the activated complex depend on the state of stress at the crack tip, and hence, are the prime determinants of the crack-velocity exponent value.[3] According to Ref. 3, the chemical potential of water does not depend on the state of stress at the crack tip and hence has no effect on the crack-velocity exponent. However, it can be supposed that the stress-enhanced dissolution of glassy phase in the water affects the composition of both the ceramics and the environment near the crack tip resulting in change of their chemical potentials and, therefore, of the crack-velocity exponent. Besides, the water concentration influences the position of stage II on the v-K diagram (Fig. 1) resulting in an increase of the mean subcritical crack-growth velocity with an increase of the water content in the environment.

Finally, it should be noted that the standards for flexure strength testing of advanced ceramics require the straining rate 0.5 mm mn^{-1} (US, ASTM C 1161 and Japan, JIS R 1601) or even 0.2 mm mn^{-1} (France, AFNOR B41-104).[11] Obviously, the results of the present study indicate the possibility of the straining-rate dependence of the measured strength values when such a low straining rate is used. Because of the different crack-velocity exponent values for different ceramics, this effect may result in non-correct strength comparative evaluations.

4 CONCLUSION

The strain-rate dependence of the strength of various alumina-base ceramics was examined in humid air and in water environments. A dynamic fatigue test method was employed with special emphasis on proper choice of the straining rates range. The following results were obtained.

1. Crack velocity exponent at the stage I of the kinetic diagram depends slightly on water content in environment, and the water content influences the mean crack-growth velocity.
2. Grain-boundary glassy phase decreases significantly the fatigue resistance of alumina-base ceramics. The effect is most pronounced for ceramics bonded with alkali-free phase.

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