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The effect of anelasticity and phase transformation on crack growth in Y-TZP ceramics

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Crack growth behavior has been investigated under monotonic and cyclic loadings for Y-TZP that produces remarkable anelastic strain. Monotonic loading testing was carried out under the condition of various stress rates (8×10^2, 8×10^{-1} and 8×10^{-4} MPa/s) and temperatures (RT and 373 K). Resistance of crack propagation was observed at the lowest stress rate at elevated temperature. Cyclic fatigue crack growth rate was examined under the condition of different frequencies and stress waveforms. Crack growth rate clearly depended on stress waveform, which was explicable by exhaustion and restoration of anelasticity at the crack tip region. Experimental results make it clear that anelasticity works as strong resistance against crack growth. In this study, the effect of environment-induced tetragonal to monoclinic phase transformation on fracture strength was also investigated for pre-cracked sample. Aged (transformed) samples have shown extreme crack closure and considerable improvement in strength. © *2001 Kluwer Academic Publishers*

1. Introduction

Transformation toughening has been established as the dominant toughening mechanism in many zirconia based ceramic materials [1–3]. Besides, some other mechanisms have been proposed and studied by many researchers since the first report on transformation toughening mechanism by Garvie *et al*. [3]. Especially, ferroelastic domain switching, detected by Virkar and Matsumoto [4, 5], has been vigorously studied as efficient mechanism at high temperature, because it is activated in place of *t–m* transformation at higher temperature. Several years ago, the authors have detected unique anelastic behavior in Y_2O_3 stabilized tetragonal zirconia polycrystalline (Y-TZP) ceramics [6, 7]. When stress is applied to this material abruptly, the strain is not simultaneously produced but gradually formed time-dependently. Subsequently, after unloading it takes some time to reach the original level [6–8]. In the previous work [9], we have investigated the relation between stress rate and fracture strength in some kinds of pre-cracked zirconia ceramics and found unique stress rate dependence in Y-TZP, indicating extremely high strength in lower stress rate region at 523 K. It is assumed that this phenomenon took place due to activated anelasticity. Therefore, anelasticity is considered to work as significant resistance against crack growth.

The study on crack growth behavior in zirconia ceramics is not systematized as thoroughly as that in other non-transformable ceramics because many toughening mechanisms might affect crack growth in a complicated way [1, 2]. In many papers the crack growth rates in zirconia ceramics are discussed in terms of *t–m* phase transformation [10–13], while there seems to be few reports paying attention to the effect of ferroelastic domain switching or anelasticity on crack growth [9]. Alcalá and Anglada [13] have investigated fatigue crack growth rate in Y-TZP at RT and 723 K and reported that cyclic crack growth velocities at 723 K were lower than prediction based upon the results of static crack growth data. This result might imply that activated ferroelastic domain switching or anelasticity influences crack growth behavior.

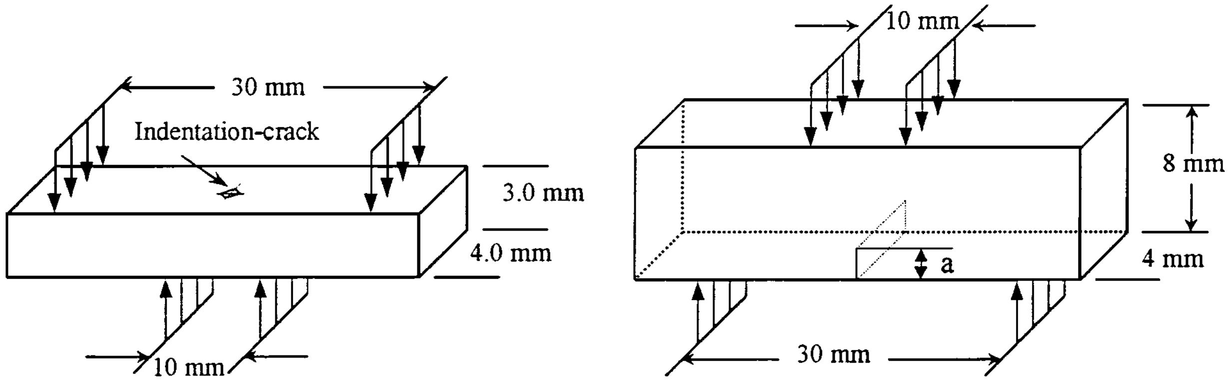
In this study, crack growth behavior under monotonic and cyclic loadings was investigated for Y-TZP ceramics. It was discussed how anelasticity and other toughening mechanisms could influence crack growth behavior.

2. Experiments

2.1. Materials

The materials prepared in this study were 2 or 3 mol% Y_2O_3 stabilized tetragonal zirconia polycrystals (2Y- or 3Y-TZP) that produces remarkable anelastic strain and 6 mol% Y_2O_3 fully stabilized zirconia (6Y-FSZ) that was considered to have no dominant strengthening-toughening mechanism.

For monotonic loading experiment, 2Y-TZP was employed. Samples were made in rectangular shapes with $3 \times 4 \times 30$ mm. Main surfaces (4×30 mm) of all the samples were polished with 1/4 μm diamond paste, then indented at the central part of their surfaces with a



(a) (b)

Figure 1 Test pieces. (a) pre-cracked specimen for monotonic loading, (b) SEPB specimen for cyclic loading.

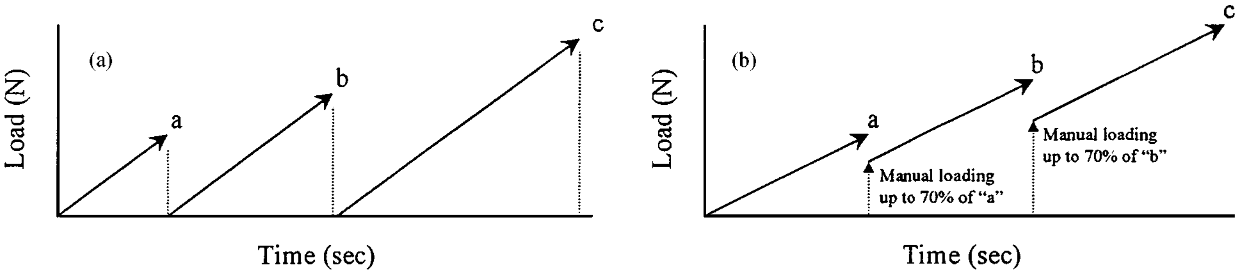


Figure 2 Procedure of load applying to measure crack length during loading process, in the case of; (a) 8×10^2 MPa/s and 8×10^{-1} MPa/s, (b) 8×10^{-4} MPa/s.

Vickers indentation load of 294 N as shown in Fig. 1a. For cyclic loading experiment, 3Y-TZP was employed and also 6Y-FSZ was used for comparison. Single edged pre-cracked beam (SEPB) specimen with dimension of $8 \times 4 \times 40$ mm was prepared for this experiment (Fig. 1b). Long through-thickness cracks were introduced by using load-bridge indentation technique.

2.2. Measurement of crack growth under monotonic loading

Testing under monotonic loading was conducted in four-point bending fixture with inner and outer spans of 10 mm and 20 mm respectively, under the condition of various stress rates (8×10^2, 8×10^{-1} and 8×10^{-4} MPa/s) and temperatures (RT and 373 K) in air. The testing at 373 K was carried out in the electric furnace fixed in loading machine and specimens were preheated for 1h before loading. The testing machine is the electrohydraulic type servopulser (EHF-EB 10kN-10L, Shimazu, Kyoto, Japan) that can execute load/strain controlled dynamic fatigue test. In order to measure crack length during loading process, load applying was carried out in the procedure shown in Fig. 2a schematically. First, loading was interrupted halfway and then the sample was taken away out of the testing machine for the measurement of crack length by optical microscope. Next, this sample was reloaded from zero-stress again up to the slightly increased stress level and crack length was measured as well. In the case of the lowest stress rate (8×10^{-4} MPa/s), sample was reloaded after loading up to 70% of latest stress level manually in order to shorten testing (loading) time, as shown in Fig. 2b.

It is well known that *t*–*m* transformation occurs on the surface of TZP materials by aging for the temperature range of 423–673 K, especially at 523 K [14–16]. In the present study, after aging the pre-cracked samples for 50h at 523 K, they were fractured by similar four-point bending system under the condition of different stress rates (8×10^1 and 8×10^{-1} MPa/s) at 523 K in air. Fracture strength measured was compared with the results of non-aged samples in our previous work [9].

2.3. Measurement of crack growth under cyclic loading

Fatigue crack propagation experiment was carried out in four-point bending fixture with inner and outer spans of 10 mm and 30 mm respectively. The testing machine was the same as that used for monotonic loading experiment. Cyclic fatigue crack growth rate was measured for the frequency range of 0.01–10 Hz (0.01 Hz (triangle wave), 0.1, 1 and 10 Hz (sine wave)) in air to study the effect of anelasticity on crack growth behavior. Stress ratio was 0.1.

For another experiment, three types of stress waveform were employed; loading time (and unloading time also) was different (9.5, 5.0 and 0.5 s) for each waveform, but total time per cycle was identical (10 s) for all, as schematically shown in Fig. 3 (each frequency corresponds to 0.1 Hz). Stress ratio adopted was 0.1. The

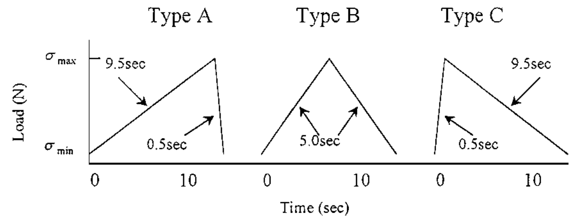


Figure 3 Schematic illustration of each stress waveform.

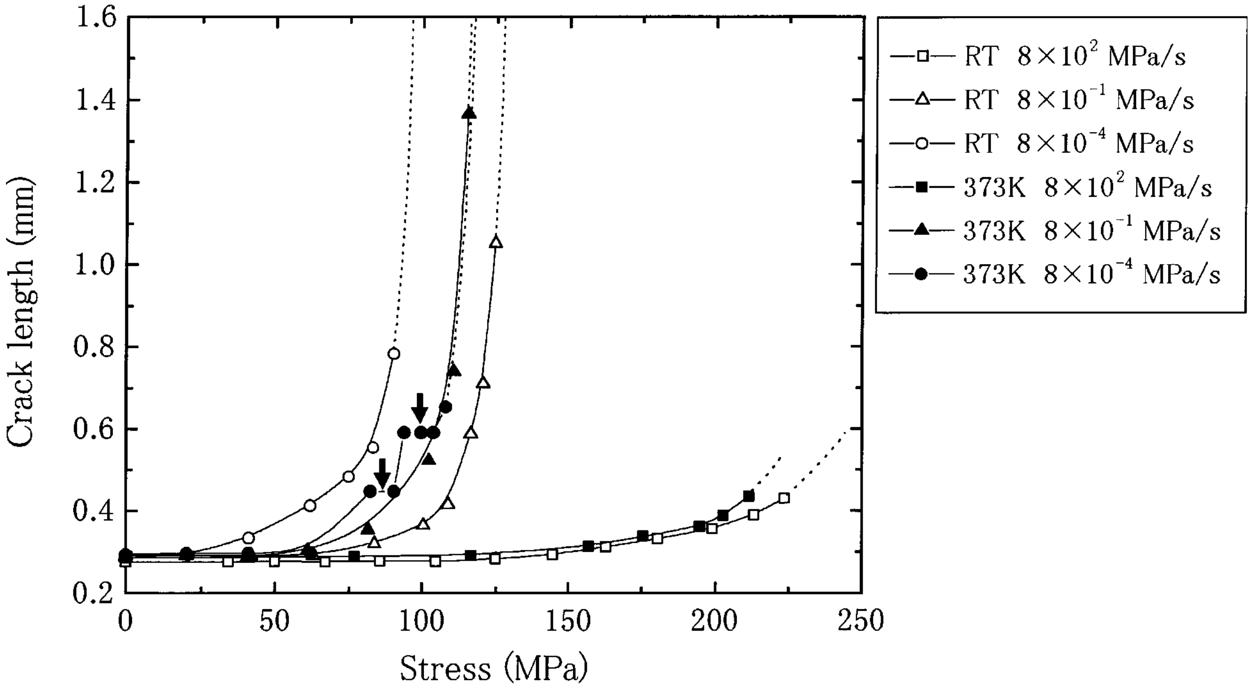


Figure 4 Crack growth behavior of 2Y-TZP during monotonic loading process. Arrows show crack arresting regions.

extent of static fatigue damage should be the same under each waveform because of the identical total time per cycle. Besides, the change of crack opening displacement (COD) with time elapse was measured by using clip-gauge.

3. Results and discussion

3.1. Crack growth behavior under monotonic loading

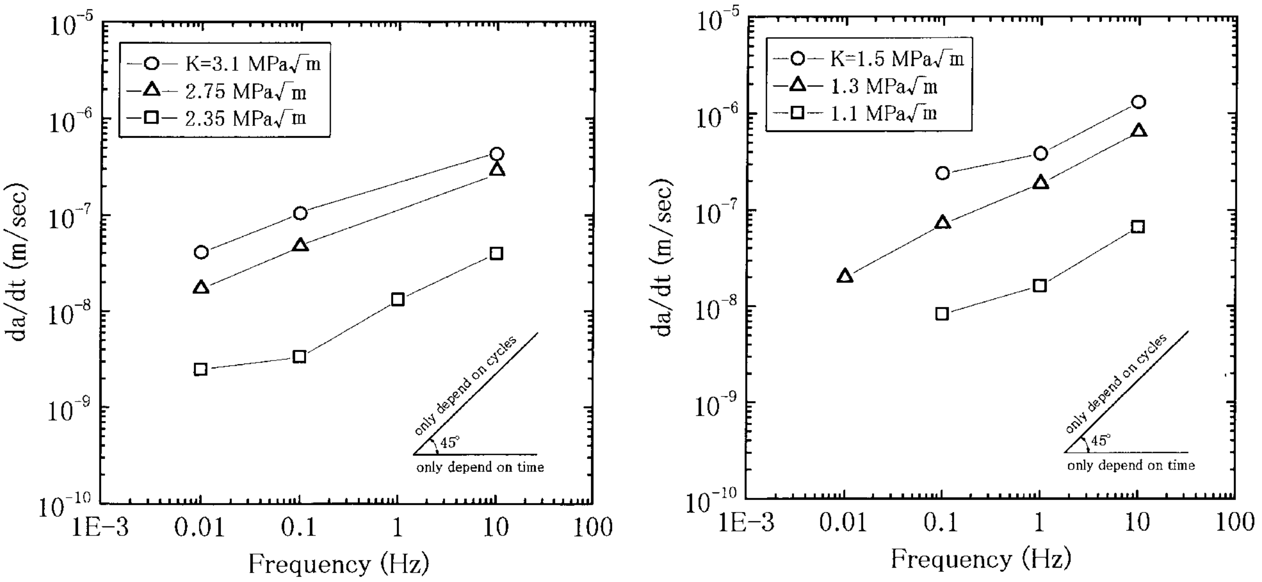
Fig. 4 shows crack growth behavior in 2Y-TZP during monotonic loading process under the condition of each temperature and stress rate. At both temperatures, slow/subcritical crack growth (SCG) occurred extremely and fracture stress decreased with decrease of stress rate. This phenomenon suggests that zirconia ceramics are so sensitive to environment. The extent of this sensitivity has been investigated in detail in our previous work [9]. It should be noticed that in the behavior under the condition of high temperature/low stress rate (373 K/8 × 10^{-4} MPa/s), resistance of crack growth was clearly observed; the first commencement of crack growth was delayed compared to RT crack behavior and a few crack arresting regions (marked by arrows in Fig. 4) were observed. Such behavior was not detected under the condition of the same stress rate at RT. Since anelasticity in Y-TZP produces non-linear strain time-dependently and is activated at the higher temperature, as seen in smooth specimens [8], it is considered that also in the crack tip region of the pre-cracked body anelastic behavior is more activated under the condition of high temperature/low stress rate and relieves the stress concentration in this region. Especially, the occurrence of crack arresting during loading process makes intensively us envisage the effect of anelasticity. It is assumed that anelastic ability could be recovered fully when loading was interrupted for the measurement of crack length. As the result, the interesting hesitation of crack growth might be observed.

There is a possibility that the above crack growth resistance was produced by environment (temperature)-induced phase transformation [14–16] causing strong crack closure effect [9] (See Section 3.5), because sample was aged for a long time during loading process. In order to check this possibility, the side surface of a sample fractured in 373 K testing was examined by using X-ray diffraction technique. However, monoclinic phase has not been detected on the side surface, which suggests that the crack growth hesitation is not concerned with environment-induced phase transformation. Besides, it is noticed that crack arresting region was not observed under the condition of the lowest stress rate/RT. Taking it into account that anelastic strain increases with temperature [8], it is considered that the amount of anelastic strain produced during loading much contributes to crack arresting observed.

Therefore, anelasticity is expected to work as strong resistance against the commencement of crack growth. Once a crack begins to propagate, however, it enters the small region exhausting the ability of anelasticity, which leads to the higher crack growth rate. In other words, anelasticity may not have extremely strong effect in the process of crack propagation. It is considered that the above two processes are repeated and the unique crack growth behavior was seen under monotonic loading condition.

3.2. Frequency dependence of cyclic crack growth rate

The relations between crack growth rate da/dt and frequency for 3Y-TZP and 6Y-FSZ are shown in Fig. 5. Since the X and Y axis of logarithms are the same scale, the line linking the plots is horizontal if the crack growth depends only on time, while its inclination is 45 degree if the crack growth depends only on the number of cycles. In 3Y-TZP, the crack growth rate decreases with decreasing frequency. The inclinations of the lines are



(a) 3Y-TZP (b) 6Y-FSZ

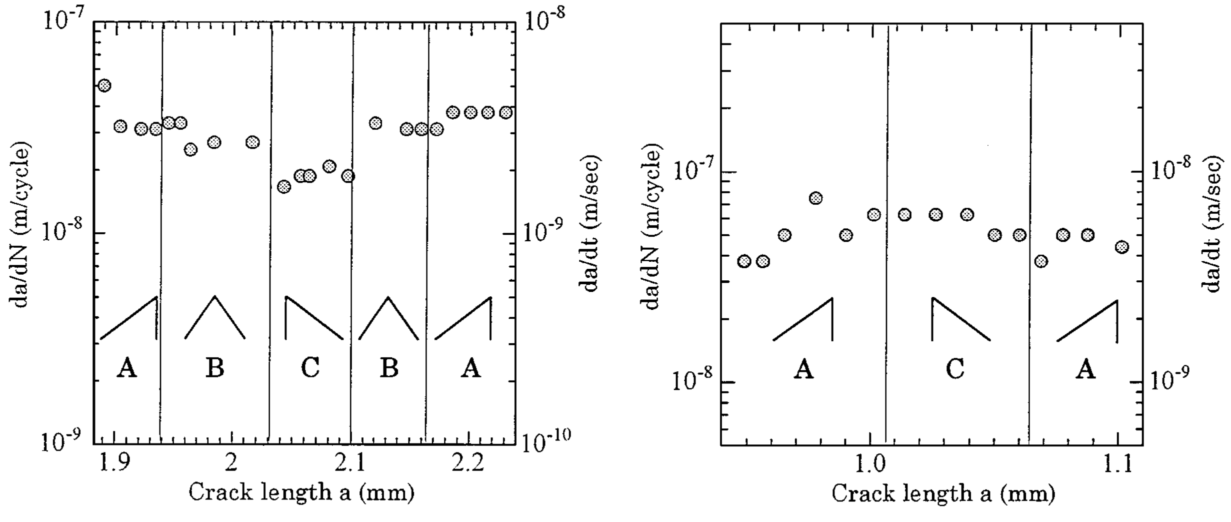
Figure 5 The relation between cyclic fatigue crack growth rate and frequency.

less than 45 degree. The crack growth rate in 6Y-FSZ also decreases with decreasing frequency similarly to that in 3Y-TZP. Such results indicate that crack growth rates in these materials are affected by both cycles and time.

Anelasticity should be more active at crack tip region at lower frequency than at higher frequency. Therefore, it is expected that anelasticity may produce frequency dependence of cyclic crack growth behavior. For 3Y-TZP, however, the data of crack growth rate at 0.01 Hz is almost on the prolonged line from the data at higher frequency, which indicates that the crack growth rate was not improved even at extremely low frequency. The same tendency is observed for 6Y-FSZ that produces lesser anelastic strain than 3Y-TZP. Anelasticity seems to have little effect on the frequency dependence of cyclic fatigue crack growth rate. With lowering frequency, not only apparent toughness increases due to occurrence of the large amount of anelastic strain but also, at the same time, the extent of brittleness due to exhaustion of anelastic strain increases in crack tip region. Therefore, it is considered that resistance and acceleration of crack growth occurs simultaneously at lower frequency and, as a result, frequency dependence is not observed for all frequency level considerably.

3.3. Stress waveform dependence of cyclic crack growth rate

Fig. 6 shows the effect of stress waveform on crack growth rate in 3Y-TZP and 6Y-FSZ. The results make it clear that crack growth rate in 3Y-TZP having intensive anelastic property strongly depends on stress waveform; the growth rate at type A (slow loading/fast unloading waveform in Fig. 3) is higher than that at type C (fast loading/slow unloading waveform in Fig. 3). In



(a) 3Y-TZP (b) 6Y-FSZ

Figure 6 The effect of stress waveform on cyclic fatigue crack growth; (a) 3Y-TZP ($K = 2.35$ MPa $\sqrt{m}$), (b) 6Y-FSZ ($K = 0.9$ MPa $\sqrt{m}$).

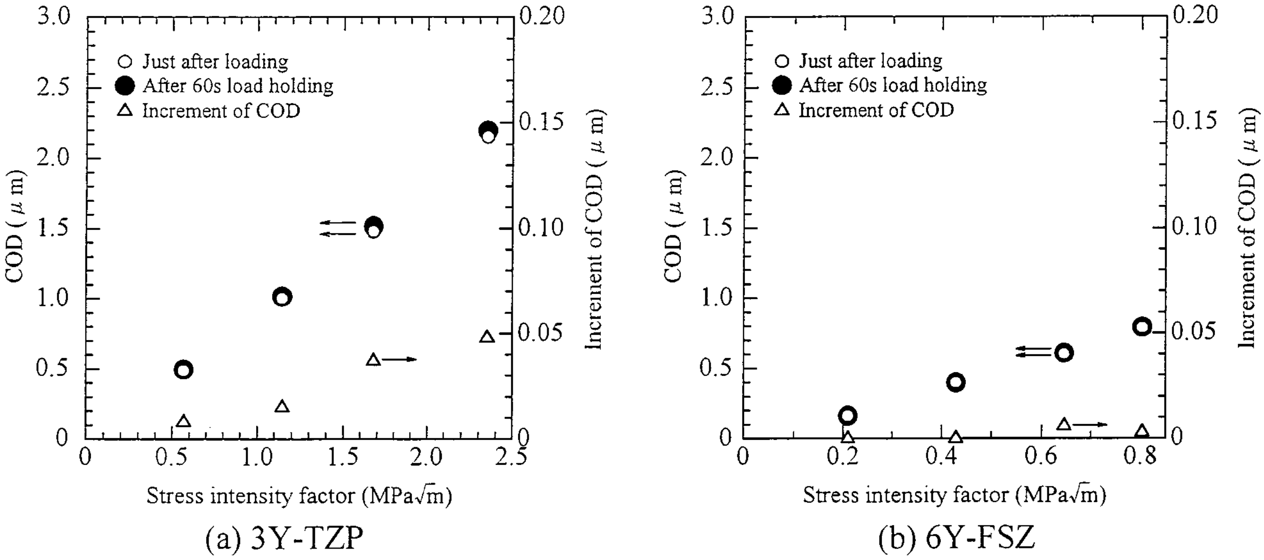


Figure 7 The change of COD caused by 60 s load holding.

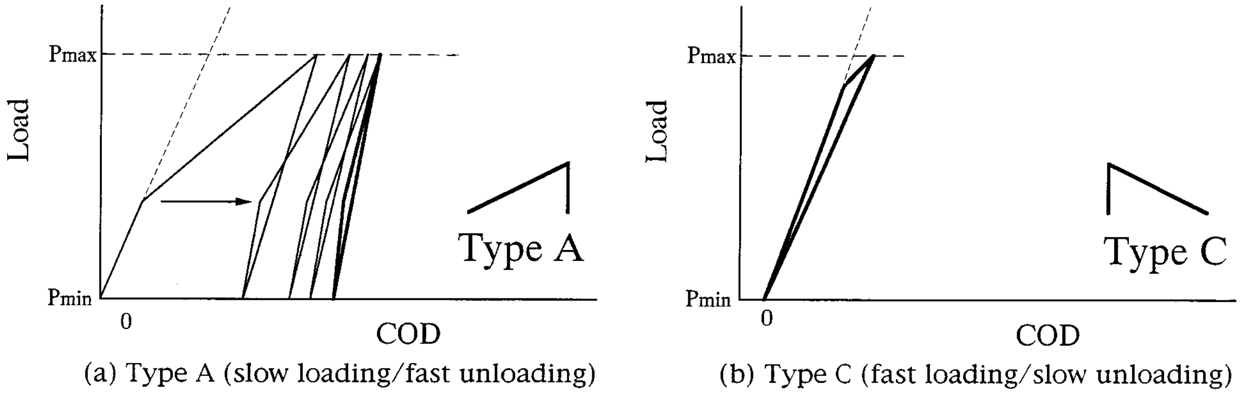


Figure 8 Schema indicating the load-displacement diagram for 3Y-TZP subjected to cyclic loading.

6Y-FSZ having lower anelastic property than 3Y-TZP, on the other hand, such a nature of stress waveform dependence was not observed. Therefore, it is assumed that anelasticity has some influence on cyclic crack opening behavior. In order to clarify the cause of this stress waveform dependence, the change of crack opening displacement (COD) with time elapse was measured under various levels of stress intensity factor. Stress was applied to the specimen and kept for 60 s at a certain stress level. In order to avoid occurrence of anelastic strain during loading, stress was applied abruptly (for 1 s). This result is shown in Fig. 7, which compares behavior in 3Y-TZP with that in 6Y-FSZ. The obvious increment of COD was observed in 3Y-TZP but not in 6Y-FSZ, which implied that crack tip blunting occurred in 3Y-TZP.

On the basis of the above result, we can schematically depict the cyclic crack tip behavior. Fig. 8 shows the load—displacement diagram for 3Y-TZP subjected to cyclic loading in two different stress waveforms of type A and C. In the case of type A, it takes a time for initial loading process so that anelasticity can work actively and the large amount of anelastic strain is produced. As the result, that crack tip region is blunted and crack propagation is much harder. Because of the subsequent fast unloading, however, specimen will be reloaded before the anelastic strain produced can recover. The state of crack tip region is schematically shown in Fig. 9a. The saturated region of anelastic strain is formed in crack tip front. Therefore, the crack tip region might be always embrittled during cyclic process in type A, leading to higher crack growth rate. On the other hand, in the case of type C, anelasticity produces anelastic strain to some limited extent during loading process, which makes crack tip blunt. Besides, since it takes a sufficient time for unloading, anelasticity could recover fully. In other words, in type C the ability of anelasticity is always operative as shown in Fig. 9b, which results in the lower crack growth rate.

It is difficult to explain this stress waveform dependence of crack growth rate by other strengthening-toughening mechanisms (stress-induced transformation [1–3, 17], ferroelastic domain switching [4, 5, 18–21], etc.), because they do not have the nature of time-dependent deformation and/or restorable strain. As for ferroelastic domain switching, for example, a domain is switched to another state by external stress higher than a certain critical level, but the switched domain cannot return to original state even though stress is removed because of existence of high energy barrier between both states [18, 19]. Kisi *et al.* [20] have detected existence of inverse switching by *in situ*

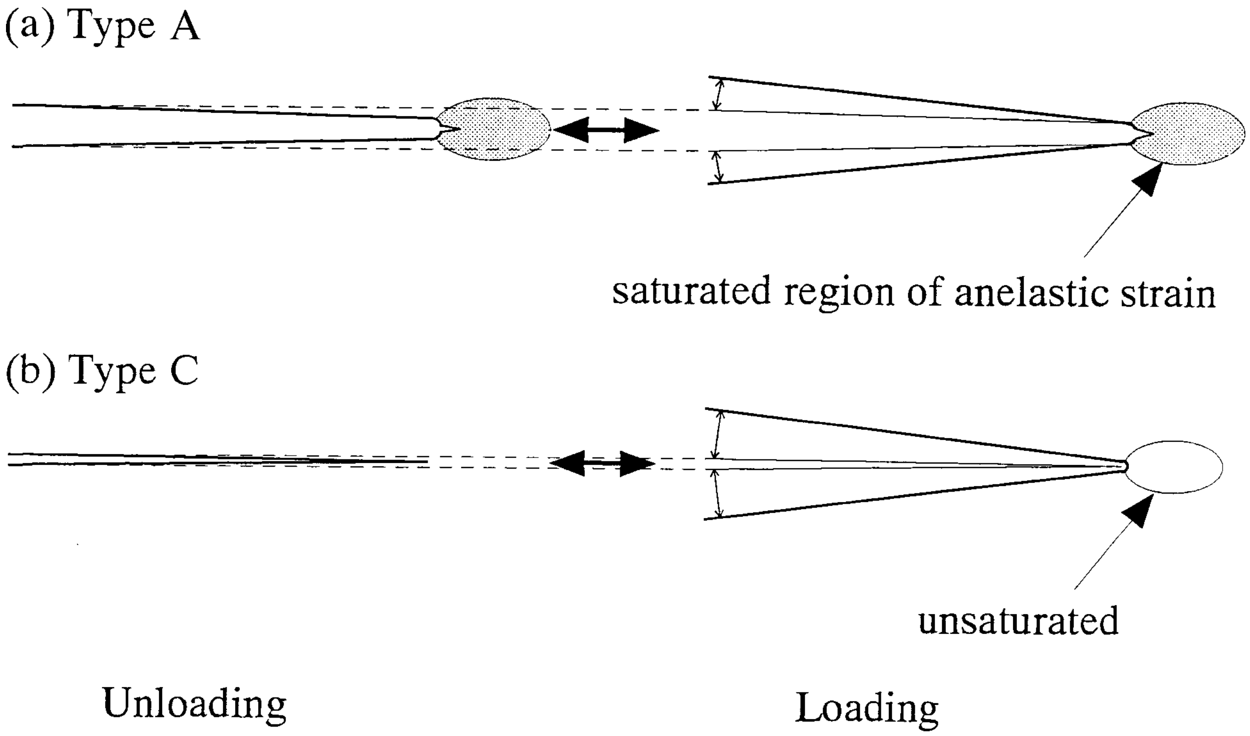


Figure 9 Schema indicating the condition of crack tip region for 3Y-TZP subjected to cyclic loading.

neutron diffraction technique. However, marvelous high stress (>1200 MPa) was required for switching and the amount of inverse switching was very small. Besides, Mehta and Virkar [21] have asserted that it takes no time for domain switching to extend through whole sample for PSZ. It is well known that *t–m* transformation also occurs in burst [17] as well as domain switching. The existence of reversible *m–t* phase transformation has been observed under relatively low stress level (as low as ≈200 MPa) by Marshall and James [22] for Mg-PSZ, but time-dependence property was not detected. Furthermore, such reversibility has not been observed for 3Y-TZP in our previous work [6]. On the other hand, anelasticity produces time dependent and recoverable strain different from above mechanisms. Therefore, we firmly believe that stress waveform dependence was caused by anelasticity and it plays an important role for cyclic fatigue crack growth behavior.

3.4. Mechanism of anelasticity and its effect on crack growth behavior

It is known that the amount of oxygen vacancy in matrix is dominated by the valence of additive cation (dopant) [23–25]. For example, the addition of Y_2O_3 into ZrO_2 replaces Zr atoms with 4 valences in zirconia by Y atoms with 3 valences. As a result, some oxygen ions and vacancies are formed in Y_2O_3 doped zirconia ceramics. On the other hand, CeO_2 doped zirconia ceramics have few vacancies because the valence of Ce atoms is the same as that of Zr atoms [23, 26]. Accordingly, in the case of Y_2O_3 doped zirconia used in this study, large amount of oxygen vacancies is involved in matrix and slight shift of ions seems to be caused by considerably low external stress. Therefore, it is assumed that anelasticity similar to elastic after-effect in metal is observable in Y-TZP ceramics. On the other hand, anelasticity has not been detected in Ce-TZP that has few oxygen vacancies [27]. Ozawa *et al.* [26] have found that Y doped zirconia had higher value of internal friction and anelastic relaxation than Ce-TZP. These results strongly make us believe that mechanism of anelasticity is concerned with the existence of oxygen vacancy irrespectively of the kind of crystallographic phase (tetragonal or cubic phase), though further study is required.

It is thought that the easiness and the range of shift of ions must increase with increase of temperature. Therefore, it is natural that anelasticity is activated under the condition of higher temperature and lower strain (stress) rate, differently from athermal *t–m* phase transformation. Anelasticity produces recoverable non-elastic strain and relieves stress concentration at the crack-tip region, resulting in crack growth resistance and increased toughness of materials. However, when anelastic strain is saturated, the ability to produce further non-elastic strain decreases, as a result, toughness decreases by contraries. After all, there is a competition between the beneficial effect due to occurrence of anelastic strain and detrimental effect due to its exhaustion.

3.5. Crack closure effect by phase transformation

It is well known that *t–m* phase transformation induced by aging at low temperatures (range of 423–673 K) will bring about strength degradation because of occurrence of microcracks on sample surface, which is introduced by such transformation accompanied by large volume expansion [14, 15]. This environment (temperature)-induced transformation is considered to be produced by the water or oxygen [14–16]. Such transformation inducing microcracks on surface has been usually regarded as harmful effect for strength of materials. For

TABLE I Comparison of fracture stresses of non- and aged samples

Stress rate (MPa/s)	Non-aged (MPa)	Aged (MPa)
8×10^{1}	160	233
8×10^{-1}	136	212

pre-cracked sample, however, in the case that transformation occurs on crack surface, volume expansion seems to introduce crack closure and work as a favorable effect. The existence of closure itself has been observed by scanning electron microscopy (SEM) in our previous work [9]. We consider that the great increase of fracture strength in the lower stress rate region at 523 K [9] was caused not only by activated anelasticity but also by this crack closure effect. Since it takes about 4 days to load to fracture at the lower stress rate, sample has been thoroughly aged and transformation has been induced during loading process.

In order to provide evidence of crack closure strengthening effect by environment-induced transformation [14–16], fracture strength was measured for pre-cracked samples aged at 523 K for 50 h prior to fracture testing. Table I shows comparison of fracture stresses of non-aged and aged samples at 523 K. (Data of non-aged samples are taken from our previous work [9]. These samples are also aged at 523 K for only 1 h prior to testing, but such short aging time does not cause transformation.) It is clearly confirmed that both strength of aged samples are considerably higher than those of non-aged by 45–55%. Considering these results, it is assumed that transformation induced by low temperature aging also works as desirable crack closure effect and makes crack propagation difficult as well as classical stress-induced transformation.

4. Conclusion

Based on the research on crack growth behavior of zirconia ceramics having various strengthening-toughening mechanisms under monotonic and cyclic loading, we conclude:

(1) Anelasticity seems to work as strong resistance against crack propagation. However, this effect decreases under the condition that anelastic strain is exhausted (saturated).

(2) Cyclic fatigue crack growth rate in Y-TZP depends on stress waveform. This is because crack tip state is affected by anelasticity effect.

(3) It is assumed that phase transformation induced by stress and by low temperature aging on crack surface promotes crack closure effect and restrains crack propagation.

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