FATIGUE BEHAVIOR OF TETRAGONAL-ZrO_2 BASED CERAMIC

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Abstract. *Cyclic fatigue strength and mechanical properties of 3mol%Y_2O_3-stabilized tetragonal zirconia polycrystalline (3Y-TZP) ceramics were studied. Samples were compacted by uniaxial pressing (50 MPa) and sintered in air at 1600 ^0C for 120 minutes. Sintered specimens were characterized by X-Ray diffraction and scanning electronic microscopy. Hardness and fracture toughness were determined using Vicker's identation method. The bending strength was determined by four-point flexure test. Highly dense sintered specimens presented hardness, fracture toughness and bending strength of 13.5 GPa, 8.1 MPa.$m^{1/2}$ and σ_f = 880 MPa, respectively. The cyclic fatigue tests were realized as four-point bending tests within a frequency of 25 Hz and stress ratio R of 0.1. The increasing of load stress lead to decreasing of the cycle number and the run-out specimens number. The t-m transformation observed through X-Ray diffraction of the fractured surfaces occurs during fracture of specimens. Samples 3Y-TZP clearly present a range of loading conditions where cyclic fatigue can be detected. The results allowed concluding that the fatigue strength limit over 5 x 10^6 stress cycles is about 550 MPa or around 63% static strength for this material. The Weibull analysis was employed in order to perform failure probability calculations.*

Keywords*: ceramics sintering, ZrO_2, mechanical properties, fatigue, Weibull probability.*

1. INTRODUCTION

Ceramic components for structural engineering applications are generally subjected to continuous operation in a variable load environment (Yao *et al.*, 2001). Usually, ceramics are characterized as regards hardness, toughness and bending strength. However, the failure under fatigue conditions, at loads much below the critical failures strength is a common phenomenon in all materials, including ceramics (Basu and Sarkar, 1992). Consequently, it is very important to make clear the fatigue strength behavior of ceramics.

The development of advanced ceramic during recent years followed several fascinating concepts, e. g., increased strength, reduced brittleness, and high temperature stability. However, the development of structural ceramics with improved fatigue resistance was not considered. The existence of cyclic fatigue effects was proved for several ceramic materials, and clear experimental evidence was reached for a limited range of test conditions (Grathwohl and Liu, 1989).

Another possibility of application for ceramic materials is on the field of biomaterials. In this case, the use of advanced ceramics started in the 1970'ies, and since then a continuous improvement of these materials in various applications can be noted. An important improvement has been possible by the use of ceramics as dental materials. They present advantages such as aesthetic, biocompatibility and chemical inertness (Hench, 1998; Willians, 1992; Hench and Wilson, 1993).

Zirconia is the most promising bio-ceramic, due to its excellent biocompatibility. The main advantages of ZrO_2 are its higher fracture strength and fracture toughness, and lower Young's Modulus (De Aza *et al.*, 2002; Stevens, 1986; Basu and Sarkar, 1996; Basu *et al.*, 2004; Piconi and Maccauro, 1999; Piconi *et al.*, 1998; Stevens, 1981). It is of common knowledge that ZrO_2 additions may increase the fracture toughness of ceramic materials. This effect is based on the tetragonal to monoclinic phase transformation of ZrO_2, accompanied by an increase of the specific volume in the order of 3-6% (De Aza *et al.*, 2002). This volume increase generates stresses in the ceramic matrix, which difficult crack propagation. When such ceramic is used for implant material such as artificial joints or dental abutment, it undergoes loading for fairly long period (Zhu *et al.*, 2004).

Cyclic fatigue of ceramics recently became a highly attractive research field for material scientists. There is a strong demand to generate design-revelant fatigue data which are required for many of the projected applications of structural ceramics. On the other hand, knowledge of fatigue in ceramics is insufficient so far and information about the correlation between microstructural parameters and fatigue properties is still missing for most ceramic systems. Besides this lack of understanding a number of fundamental questions still have not been answered unambiguously for many of the most important ceramics (Grathwohl and Liu, 1991a; Grathwohl and Liu, 1991b).

It is essential in the engineering applications of ceramic materials for structural purposes to determine the fatigue behavior under appropriated loading, such as static or cyclic. A considerable number of reports have been published on the fatigue of glass, alumina and zirconia ceramics. There have been few critical studies published regarding cyclic and static fatigue at room temperature of advanced ceramics, although activity on this field has recently been increasing. Furthermore, fatigue testing applied to brittle materials imposes a number of problems. One of them is the wide scatter in data, which sometimes obscures the fatigue tendency. This scatter is considered to derive intrinsically from a defect distribution in the specimens (Kawakubo and Komeya, 1987).

This research is focused on the processing, mechanical properties and cyclic fatigue life of Y_2O_3 stabilized ZrO_2 ceramic. Specifically, the fatigue behavior of tetragonal zirconia polycrystals (TZP) with 3mol% of Y_2O_3 stabilized (3Y-TZP), produced by solid-state sintering at 1600^0 C was investigated by means of cyclic four-point bending load controlled tests. The occurrence of the t-m transformation during the fatigue tests was observed.

2. EXPERIMENTAL

2.1. Processing

High-purity Y-TZP commercial zirconia (ZrO_2-3mol.%Y_2O_3 - Tosoh Grade 3YSB - Japan) with 10% of residual monoclinic phase was used as starting powder. The zirconia powder was cold-pressed into matrix plates with 114 x 25 mm of measurements, under a nominal pressure of 50 MPa, by 30 seconds. Approximately 65 g of powder were used in each sample. The green compacts were sintered in air at 1600 ^oC, in a $MoSi_2$ furnace for 2 h, with heating rate of 10^oC/min up to 1100^oC; 5^oC/min up to 1400^oC; and 3^oC/min until the final temperature and cooling rate of 10 ^oC/min (Willians, 1992; Piconi and Maccauro, 1999; Piconi *et al.*, 1998).

In order to obtain the specimens for the four-point flexure tests, the ceramic plates, with post shrinkage measures of 90 x 22 x 6 mm after sintering, were cut and grinded in rectangular bars of 3 x 4 x 45 mm, according to Fig. 1, with an automatic grinder. After milling, the samples were polished with diamond paste in sequence of 15, 9, 6, 3 e 1 μm.

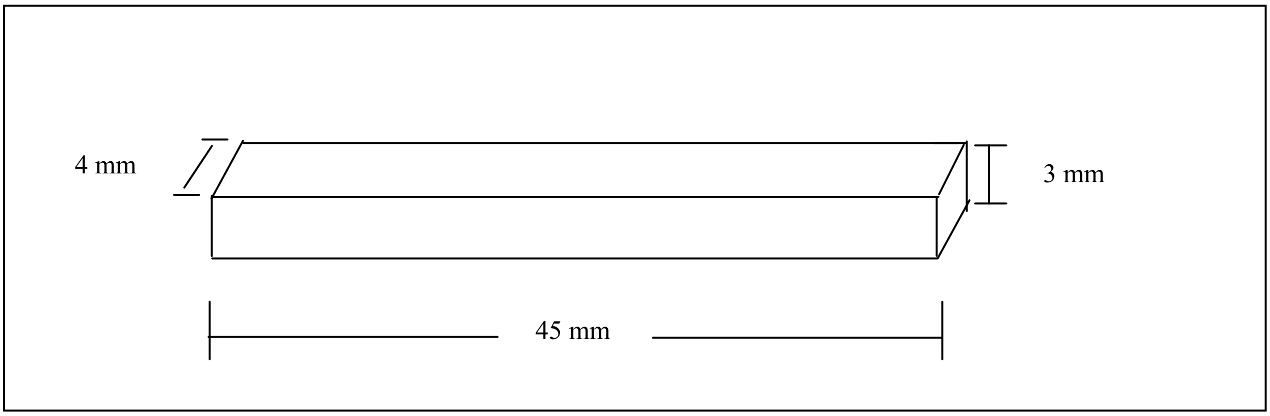


Figure 1. Fatigue and bending test specimens

2.2. Characterization

The bulk density was measured by the Archimedes' method in distilled water. The crystalline phase content was determined X-ray diffractometry (XRD) using Cu-kα radiation in the 2θ range of 20 to 80°, with a step width of 0.05° and 2s of exposure time per position. The monoclinic phase fraction was calculated using the Garvie and Nicholson (1972) method. Consequently, the monoclinic volume fraction was then obtained using equation proposed by Toraya *et al*. (1984).

Polished-thermal etched, at 1300^oC for 15 minutes, surface of the sintered samples and fractured surface of the mechanically tested specimens, were examined by scanning electron microscopy (SEM), using a LEO-1450VP microscope.

2.3. Microhardness and fracture toughness

Microhardness and fracture toughness, K_{IC}, were determined using the Vickers Indentation method. For statistical reasons, 21 indentations per sample were done, under a load of 20N, for 30s. The fracture toughness was calculated by measurement of the cracks length, using the relation proposed by Evans and Charles (1976), valid for Palmqvist cracks.

2.4. Modulus of Rupture (MOR)

The Modulus of Rupture (MOR) was determined by four-point flexure tests, using a servo-hydraulic testing machine MTS model 810.23M. For the accomplishment of the bending tests, batches of 21 samples were grinded and polished, obtaining bars of 4 x 3 x 45 mm, according ASTM C 1116 - 94. The tests were conducted using a four-point bending device with outer and inner spans (I_1 and I_2) of 40 and 20 mm respectively, as shown in Fig. 2. The speed of the crosshead displacement was 0.5 mm/s. The bending strength of the samples was calculated by Eq. (1).

$$\sigma_f = \frac{3}{2} F_A \times \frac{(I_1 - I_2)}{b \times h^2} \qquad (1)$$

Where:
σ_f – Bending strength [MPa]
F_a – Rupture load [N]
b – Width of the samples [mm]
h - Height of the samples [mm]
I_1 – Outer span [mm]
I_2 – Inner span [mm]

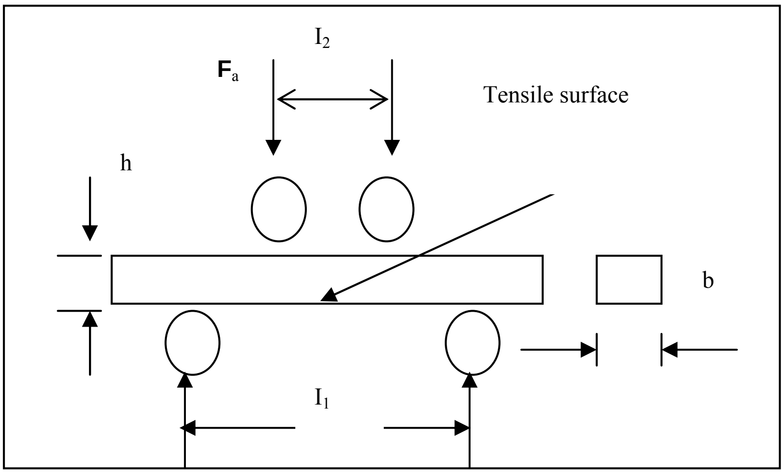


Figure 2. Schematic illustration of the 4-point bending test, with b – sample width [mm], h – sample height [mm], I_1 – outer span distance [mm], I_2 – inner span distance [mm].

2.5. Cyclic fatigue by four-point bending testing

The cyclic fatigue tests were carried out by four-point bending loading in air at room temperature, with relative humidity near to 60%. The specimen dimensions and the testing machine were the same employed in the bending strength tests. The cyclic fatigue was studied under a sinusoidal stress wave form within a frequency value constant of 25Hz and constant stress ratio (R) between the minimum stress (σ_{min}) and maximum stress (σ_{max}) was of 0.1. The number of specimens used in fatigue tests varied between 13 and 23 samples, under stresses of 570, 610 and 650MPa. In the low stress levels, 500 and 530 MPa, only 3 specimens were tested. The tests were interrupted when the survival samples reached a number of cycles between 2 and 5 x 10^6 cycles.

2.6. Statistical analysis

For the statistical evaluation of the fracture strength and cyclic fatigue resistance, the two-parameter Weibull distribution function, given by Eq. (2), was used.

$$P(x) = 1 - \exp\left(-\left(\frac{x}{b}\right)^m\right) \qquad \text{for } x > 0 \text{ e} \qquad (2)$$

$$P(x) = 0 \qquad \text{for } x \leq 0$$

Where:
P – probability associated to x value (or Failure probability)
m –modulus of Weibull distribution
b – scale parameter or characteristic strength
x – bending strength (static tests) or the number of cycles to failure (fatigue tests)

The Weibull parameters *m* and *b*, are obtained by the linearization of Eq. (2), see Eq. (3), and plotting ln ln [1/(1-P(x))] vs ln x.

$$\ln\ln\left(\frac{1}{1-P(x)}\right) = m\ln(x) - m\ln(b) \tag{3}$$

The stress value for 50% of rupture probability was estimated as reference and also for direct comparison with the average fracture stress. The Weibull parameter "*m*" was determined using a factor of correction of 0.938, corresponding to 21 samples, in agreement with the German norm DIN-51-110.

3. RESULTS AND DISCUSSION

3.1. Characterization

Figure 3 presents X-ray diffractogram patterns of ZrO_2 sample: a) before sintering, b) after sintering at 1600°C and c) in the fracture surface after bending fatigue testing.

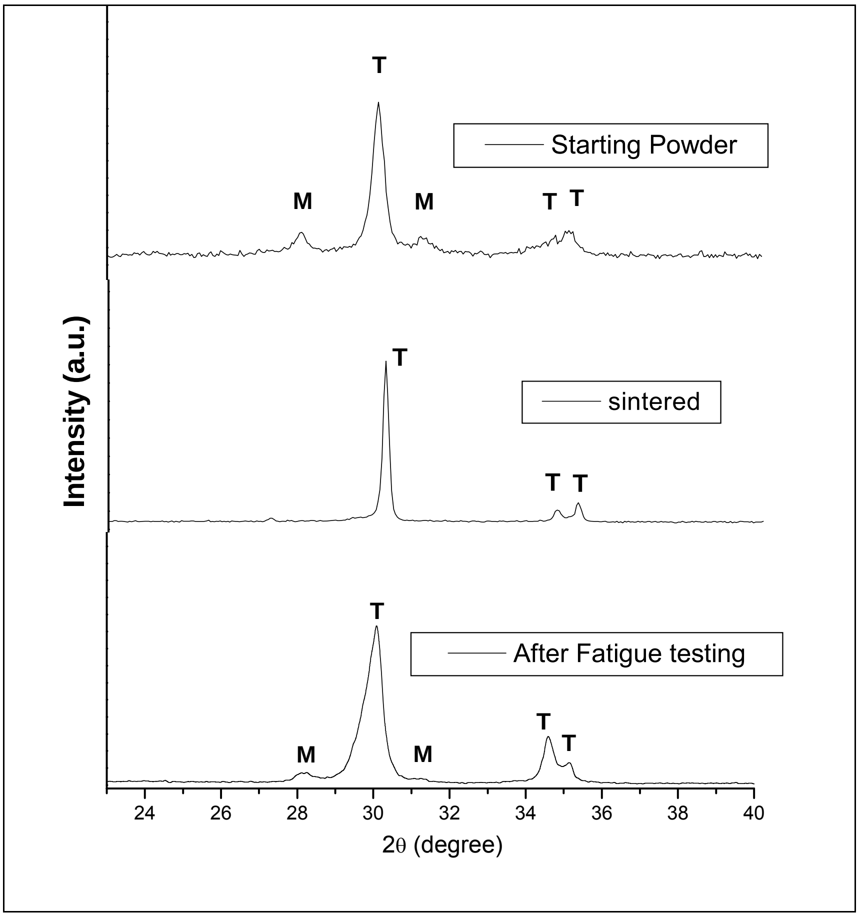
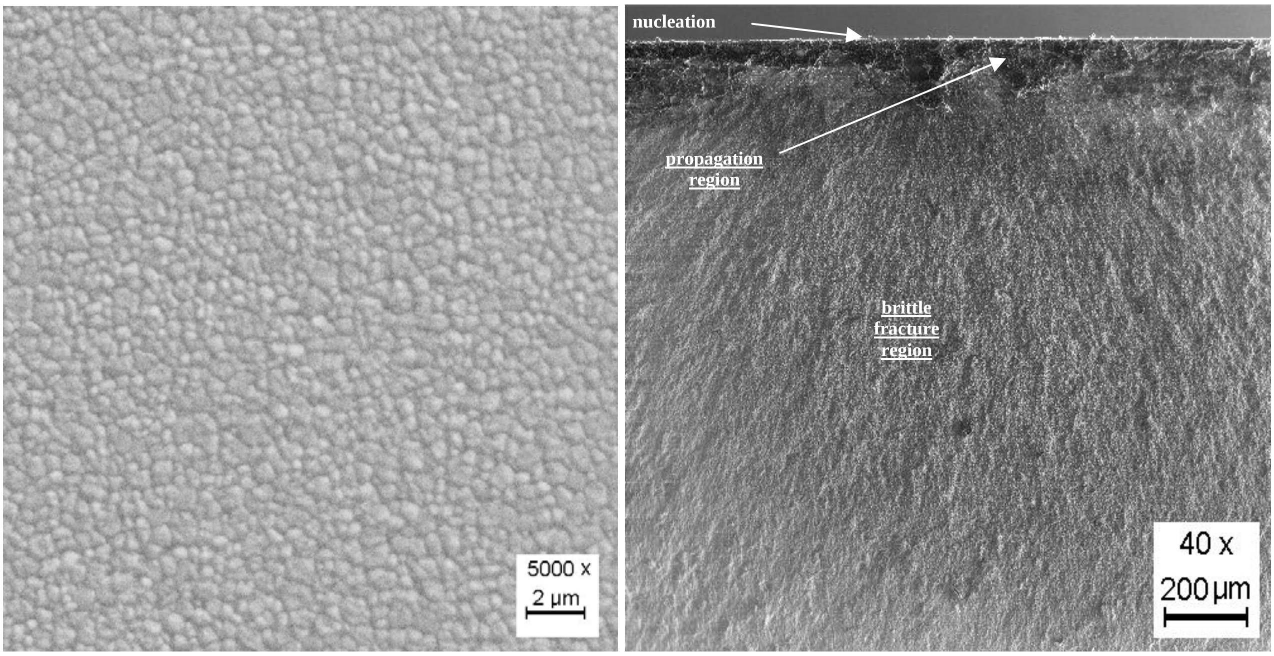


Figure 3. X-ray diffractogram patterns: a) before sintering, b) after sintering at 1600°C and c) in the fracture surface after bending testing. (T - Tetragonal; M - Monoclinic).

It can be observed that the starting powder, as received, presents a certain amount of residual monoclinic ZrO_2 phase, which was estimated, according to Toraya *et al*. (1984), in 15% vol. After sintering, the processing conditions used in this work allowed a total stabilization of tetragonal ZrO_2 phase, during cooling, by incorporation of Y_2O_3 in the structure. The X-ray diffractogram patterns of the fracture surfaces of the fatigue specimens, show clearly that there was a considerable amount of stress-induced *t-m* transformation, around 10 vol.%, due to the tensile stress to which the grains of these surfaces were submitted. It is known that the application of stresses to tetragonal ZrO_2 grains can start the martensitic transformation (tetragonal-monoclinic, *t-m*) (Willians, 1992).

Figure 4 presents a micrography of the polished/etched ZrO_2 surface and the fracture surface of a specimen tested in fatigue. Looking at the typical microstructure of sintered samples at 1600 ^oC, Fig. 2(a), it can be observed the presence of refined microstructure with equiaxial grains of mean size smaller than 0.5 µm. It was not observed the abnormal growth of ZrO_2 grains in this material. In Fig. 2(b) it is seen a typical brittle fracture surface which shows clearly that the initial crack nucleation and propagation region is located in the upper side of the picture, corresponding to maximum tensile stress occurred in the bending test.



(a) (b)

Figure 4. Micrograph of the ZrO_2 samples: a) polished surface, b) fracture surface.

3.2. Microhardness, fracture toughness and modulus of rupture (MOR)

Table 1 presents the relative density and mechanical properties of the sintered samples.

Table 1. Results of the sintered samples.

Relative density (%)	Vickers Hardness (GPa)	Fracture Toughness ($MPam^{1/2}$)	MOR (MPa)
99.7±0.2	13.5±0.2	8.15± 0.25	880±35

High densification (> 99.5), was observed in the sintered samples, indicating that the sintering conditions used in this work were satisfactory to elimination of the porosity, maintaining microstructure with refined grains, see Fig. 2(a). This typical microstructure and the results of the X-ray diffractogram patterns, showed in Figure 1(b), which indicated predominance total of tetragonal phase, justify the high K_{IC} and MOR values presented in Tab. 1. *Crack deflection* and *toughening by phase transformation* are the main mechanisms actuating to improve the mechanical properties of this material (Fett and Munz, 1996). The *t-m* phase transformation, allowed the MOR to reach the elevated values, near to 900 MPa. Furthermore, the hardness values were about 13.5 GPa. Results obtained in a previous work (Grathwohl and Liu, 1991) for ZrO_2-3%mol Y_2O_3, show hardness, fracture toughness and modulus of rupture of 12.3 GPa, 4.8 $MPam^{1/2}$ and 656 MPa respectively, but in general, the results obtained in this work are typical and consistent with literature data (Stevens, 1986).

3.3. Cyclic fatigue

The results of the cyclic fatigue tests are shown in Fig. 5 in terms of the σ x N curve which correlates the maximum bending stress level (σ_{max}) to the number of cycles to failure (N_f).

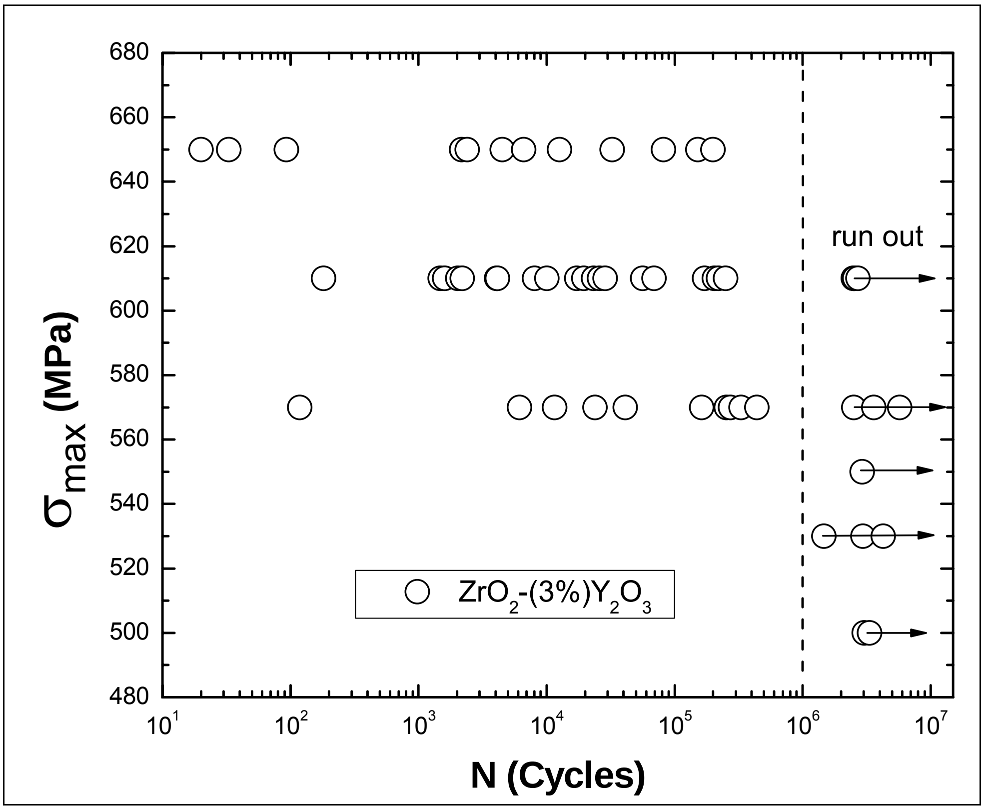


Figure 5. Cyclic fatigue tests results of the ZrO_2 samples: σ_{max} x N curves.

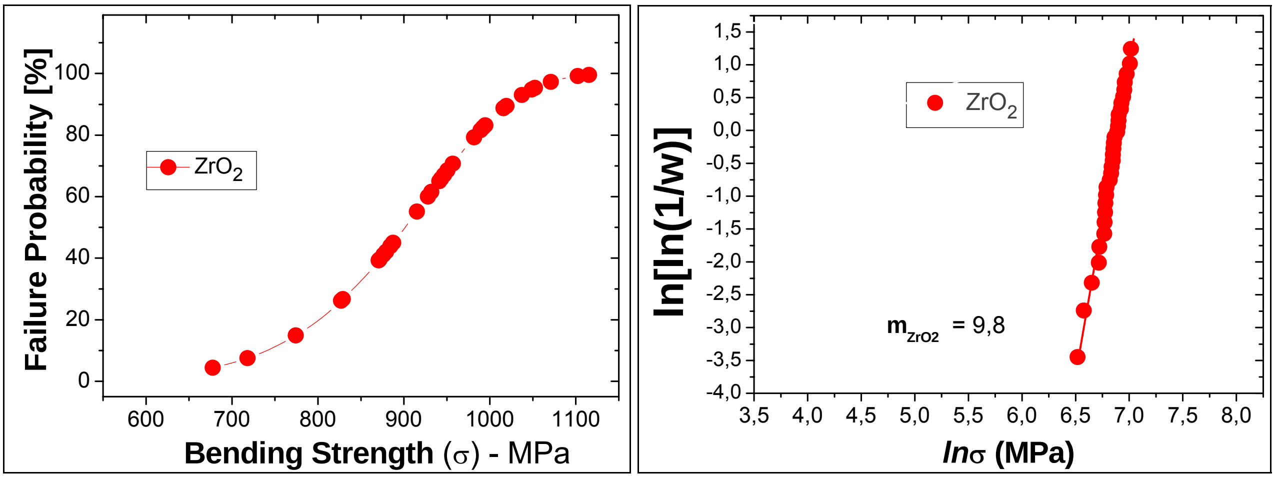
The cyclic fatigue tests were interrupted at $N_f = 2$ to 5×10^6 cycles, if failure didn't occur. The specimens which did not fracture in the test are marked by an arrow symbol (run out). The five maximum stress levels (σ_{max}) were selected in relation to the initial strength. It was found that the fatigue strength limit is around of 550 MPa, which corresponds to 62.5 % of the MOR.

At the lowest stress levels (σ_{max} = 500 MPa and 530 MPa), neither spontaneous nor fatigue fracture were observed for all specimens. As σ_{max} increased, some specimens reached $N_f = 2 \times 10^6$ cycles without failure, but some specimens failed spontaneously, i. e., below 10^3 cycles of load. On the other hand, the number of specimens failing at $10^3 < N_f < 2 \times 10^6$ was relatively large. In an amount of 13 specimens tested at σ_{max} = 650 MPa, 4 specimens failed below a hundred cycles, 9 failed during cycling, none of them achieved 10^6 cycles. The 23 specimens tested at σ_{max} = 610 MPa revealed the following: 1 specimen failed below a hundred cycles, 19 failed during cycling, 3 specimens survived 2×10^6 cycles. The 13 specimens tested at σ_{max} = 570 MPa revealed that 1 specimen failed below hundred cycles, 9 failed during cycling, 3 specimens survived 2×10^6 cycles. At the stress levels above 550 MPa, the majority of the specimens failed in the range of $10^3 < N_f < 2 \times 10^6$ cycles. Samples that failed with low cycle ($N_f < 10^3$ cycles), are more representative of bigger stresses, while the reduction in maximum stress applied lead to a significant increase in the number of unfailed samples ($N_f > 2 \times 10^6$ cycles).

The X-ray diffractogram pattern and fracture surface analysis by SEM of specimens undergone to fatigue tests are similar to the presented in the Fig. 1(c) and Fig. 2(b), respectively. The specimens rupture in fatigue begin in the polished tensile surface of the sample and occurs in brittle mode, being a function of the critical flaw size. This flaw, in turn, to initiate the propagation it suffers compression due to martensitic transformation with associated volumetric expansion increasing the crack propagation strength, until it reaches a critical value, above which the crack propagates in catastrophic mode. The mechanism of the stress induced *t-m* transformation was also observed by Grathwohl and Liu (1991). Cyclic testing of Y-TZP provides interesting results concerning fatigue behavior, threshold phenomena, and the strengthening effect of this transformation-toughened ceramic. The range of fatigue is not clearly delimited and it is clear that this fine-grained ceramic is particularly prone to cyclic fatigue.

3.4. Weibull distribution

The results of the statistical analysis of the MOR using the two-parameter Weibull approach are presented in Fig. 6.



(a) (b)

Figure 6. Failure probability and Weibull diagram of the pure ZrO_2.

It was observed for a large amount of ceramic materials that the "*m*" value depends on processing, the amount of inclusions, microstructure, pore distribution and surface finishing degree. These values are usually between 3 and 15 for ceramics, which means that materials with *m*=15 have a lower spreading of fracture strength values than ceramics with *m*=3. High values of m represent less scattering of the fracture strength and thus more reliable materials. Quinn (1991) says that groups of ceramic materials with *m* higher than 10 can be considered good and reliable for structural applications. In this work, a Weibull modulus *m*=9.8 was found for the ZrO_2 samples. The predominant factors for this resistance behavior are the microstructural characteristics and the high relative density.

Figure 7 and Tab. 2 present results of Weibull analysis of the samples tested in fatigue, in which the spontaneous failure data ($N \leq 10^2$ cycles) were excluded of the calculations. The very low *m* values reflect the high scattering of the fatigue data. A tendency is observed of the *m* values to increase and the *b* values to decrease as σ_{max} is increased.

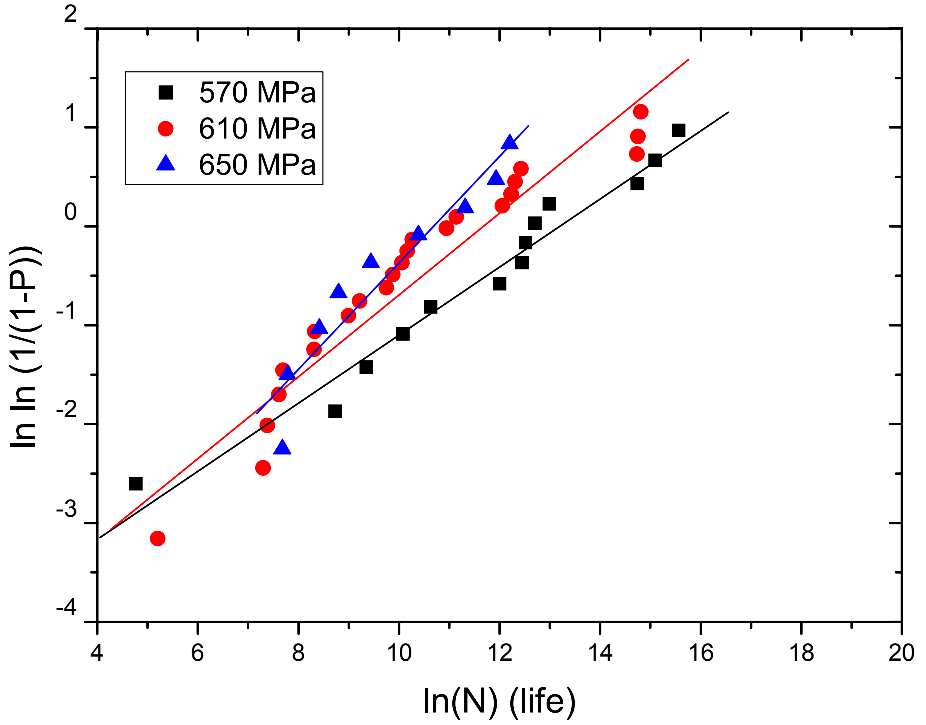


Figure 7. Weibull diagram of the of the pure ZrO_2 tested in fatigue

Table 2. Fatigue Weibull parameters.

σ_{max} [MPa]	*m*	*b* [cycles]
650	0,536	43863
610	0,413	100779
570	0,344	507194

4. CONCLUISIONS

In this work, the mechanical properties and cyclic fatigue behavior of 3mol%Y2O3-stabilized tetragonal zirconia polycrystalline (3Y-TZP) were studied.

The adopted processing route allowed obtaining high densification values (99.7%) for the sintered material. The modulus of rupture (880 MPa), Vickers hardness (13.5 GPa) and fracture toughness (8.15 $MPa.m^{1/2}$), as well as the Weibull modulus (9.8) presented by the tested samples qualify this material for structural applications.

Fatigue tests by four-point bending were conducted in order to obtain the σ x N curve for the material. The experimental results clearly indicate that 3Y-TZP ceramic material suffered cyclic fatigue fracture process. X-ray diffractometry results indicate that the material suffers t-m phase transformation due to cyclic as well as static loading. It was found that the fatigue strength limit is around 550 MPa (62.5 % of the MOR). The Weibull analysis of the results obtained low m values, reflecting the high scattering of the fatigue data.

5. ACKNOWLEDGEMENTS

The authors acknowledge to the *FAPESP* for financial support, under Grants n°. 04/04386-1 and 05/52971-3.

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