Weibull characterization of the flexural strength of hydrothermally degraded 3Y-TZP zirconia

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The flexural strength of 3% mol yttria-stabilized tetragonal polycrystalline zirconia (3Y-TZP) with a ~0.30μm grain size was studied under 4-point bending test. The material was evaluated both as received and after water vapour exposure for long times of 100hr and 200hr at 131ºC in autoclave. Results were systematically analyzed by Weibull statistics and fractography. It was found that the mean flexural strength and Weibull modulus change from 960MPa and m=10.9 in the as received condition to 916MPa (m=19.5) and 860MPa (m=19.8) for 100 and 200 hours ageing, respectively. This increase in m can be explained by a change in the origin of fracture from natural flaws in the original material to original defects in the degraded surface layer which extend under loading to a depth equal to its thickness. This layer can be clearly appreciated during fractographic analysis after long degradation times. In the present case the thickness of the degraded layer was on average equal to 8.5μm after 100 hours of hydrothermal exposure.

INTRODUCTION

Tetragonal zirconia polycrystals stabilized with 3% molar yttria (3Y-TZP) are used in hip prosthesis and in dental restoration because of their high mechanical strength (~1000MPa), wear resistance, excellent biocompatibility and moderate fracture toughness (~5MPam$^{1/2}$), which is caused by a mechanism of phase transformation toughening from the metastable tetragonal to the monoclinic crystal structure. However, this transformation can as well spontaneously initiate at the surface during hydrothermal degradation at relatively low temperature, as first observed by Kobayashi et al.[1]. This spontaneous transformation extends into the bulk and is accompanied by the formation of micro-cracks, resulting in a loss of mechanical integrity after a long exposure time. This phenomenon is frequently referred to as hydrothermal degradation HD (see Lawson [2] and Chevalier et al.[3]). In recent years, it has been shown that HD may take place in femoral heads made of 3Y-TZP at human body temperature in the presence of body fluids. Although HD may not practically change the strength after several years in vivo, it may produce roughening of the surface by phase transformation and grain pull out under contact loading. Currently, it represents a serious factor to consider for the use of 3Y-TZP in implants. The aim of the present work is to characterize, by using Weibull analysis, the influence of HD on the flexural strength of 3Y-TZP during the initial stage when the drop in the strength begins to be apparent and for whose characterization a Weibull analysis is necessary because of the large scatter in the results.

EXPERIMENTAL

The as-received material consisted in cylindrical, polished 3Y-TZP rods (*D*=7.99mm diam., 63.1mm long) delivered by Kyocera Corp. with an average grain size of 0.30µm and a density over 99% the theoretical value. Three conditions of the material were considered: as-received, and exposed to 100 and 200 hours in autoclave at 131ºC under 2bar of pressure. This accelerated degradation test was chosen because it has been reported[4] that steam sterilization at 134ºC in autoclave for 5 hours simulates the degradation that would take place after 15-20 years in vivo. For each condition, 32 specimens were broken by incremental load under a 4-point bending test, with support and inner spans of *L*=60mm and L_i=30mm respectively and a loading speed of 200N/s. For a cylindrical bar breaking under a certain load *F*, the fracture strength was estimated as:

$$\sigma = \frac{8F(L-L_i)}{\pi D^3} \qquad (1)$$

The experimental fracture data was analysed in accordance with ASTM standard 1239-00 [6]. Thus, the Weibull modulus *m* was estimated for each data set with the Maximum Likelihood Method. Further fractographic analysis of the broken surfaces was carried out by scanning electron microscopy (SEM), and the thickness of the monoclinic layer was measured by laser confocal microscopy (Olympus, LEXT OLS 3100) after polishing (up to colloidal silica finish) the cross-section of degraded specimens. Finally, it was possible to estimate the critical defect size, *a*, from the average strength of each data set, using the relationship:

$$K_{IC} = Y\sigma\sqrt{a} \qquad (2)$$

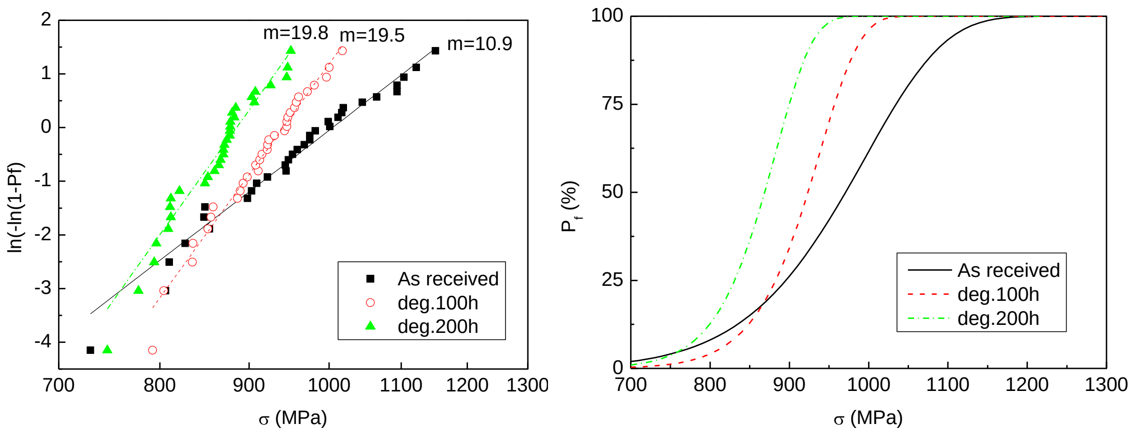
where *Y* is a geometrical factor taken here as *Y*~1.29 and K_{IC}=5.0MPam$^{1/2}$ is the tetragonal fracture toughness of our ceramic.

RESULTS AND DISCUSSION

Figure 1 shows the failure probability measurements for the conditions considered: as received and after 100 and 200 hours of hydrothermal degradation. It can be seen how the flexural strength decreases after degradation. In the as received condition, the average fracture resistance is 960MPa with a standard deviation of 101MPa. For this condition, around 40% of the specimens broke at a stress higher than 1000MPa. From this data set, the Weibull modulus of the as received ceramic was calculated as *m*=10.9.

After 100 hours of degradation, the average flexural strength decayed to 916MPa and only two specimens broke over 1000MPa, while the probability of fracture below 900MPa increased to 33%. At the same time, the corresponding degraded layer of these specimens was observable on the broken surfaces and its thickness, *d*, was 8.5µm. There was less scatter in the strength for this degraded material (st.dev.=56MPa) and the Weibull modulus thus had increased to *m*=19.5. After degradation for 200 hours, the average flexural strength decayed still more to 862MPa (st.dev.=50MPa). For this condition, no specimens broke over 1000MPa, the probability of breaking below 900MPa was 79% and the Weibull modulus was *m*=19.8.

FIGURE 1: Left: failure probability Pf measurements for the three conditions considered: as received, 100hr degraded and 200hr degraded. Right: corresponding Weibull distribution for each condition.



All these results indicate that the distribution of typical defect size present in the surface under bending becomes more homogeneous after HD. Let us assume that the critical defects in the as received conditions are semicircular surface cracks.

FIGURE 2: Left: average flexural strength (plus std. dev.) of 3Y-TZP after exposure to 131ºC water vapour in autoclave. Right: Fracture surface of a specimen degraded during 100hr.

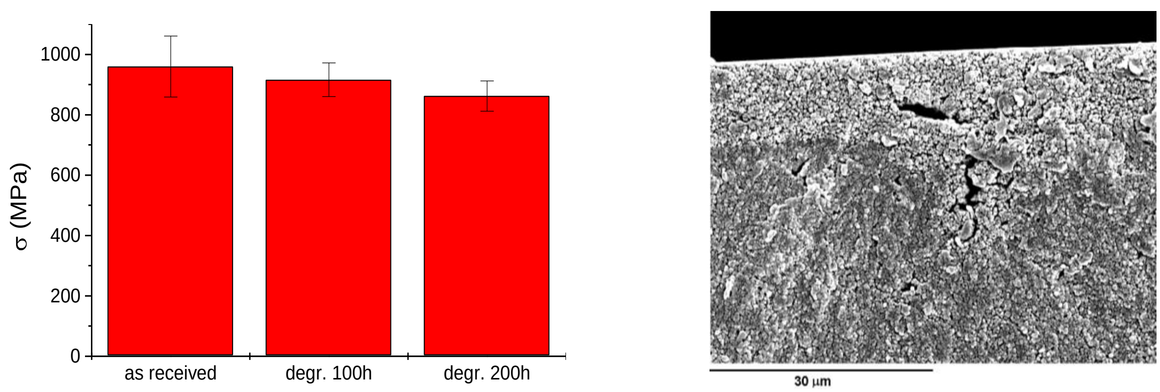


Figure 2 summarizes the observed decay in flexural strength after degradation. The estimated average critical defect size in the surface under tension in the as received condition was ~17.4μm. However, in the degraded material it is possible that fracture is controlled by cracks which extend in the degraded layer and which are stopped at the monoclinic-tetragonal interphase, because K_{Ic} for m-phase is much lower than for t-phase (about 40%). Then, original semicircular cracks will easily extend under stress on the m-surface layer in a stable manner before reaching conditions of unstable fracture at the deepest point in the t-phase, and therefore the shape factor of an elliptical crack, a/c, will decrease as the crack extends at the surface increasing in this way K_I at the deepest point. The overall effect will be to decrease the strength, but at the same time diminishing the scatter since HD will tend to homogenize the critical crack length of cracks grown inside the degraded layer. For example, in the case of a degraded layer of 8.9 μm in thickness a/c would be expected to change from 1 to about 0.13 for the critical crack and it could explain the strength and the reduction of scatter. The SEM image in figure 3 shows the fracture surface for a 100hour degraded specimen and the thickness of the degradation layer (about 8.5 μm on average) coincides with that expected in order to explain the results. Two zones can be appreciated. The observed contrast in the degraded layer indicates that its fracture mechanism is different from the bulk, probably due to the intergranular microcracking inside the degraded layer.

CONCLUSION

The flexural strength of 3% mol yttria-stabilized tetragonal polycrystalline zirconia (3Y-TZP) with a ~0.30μm grain size was studied under 4-point bending in the as received (annealed) condition and after water vapour exposure for long times of 100hr and 200hr at 131ºC in autoclave. It was found that the mean flexural strength decreases slightly and the Weibull modulus increases sharply as a result of severe hydrothermal test in autoclave. These effects are explained by the influence of a surface degraded layer that increases stable crack extension at the surface before unstable fracture starts in the tetragonal phase.

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