Fracture toughness and microstructure degradation of Y-TZP in aqueous physiological environment

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The present work aims at the study of possible time-dependent changes on the fracture toughness and microstructure of two Y-TZP ceramics with different yttria contents, when immersed in Ringer's solution. The effects of yttria content, sintering time and temperature and immersion time and temperature on the tetragonal phase content and microstructure of these ceramics were studied, using X-ray diffraction (XRD) analysis and scanning electron microscope analysis (SEM) of the sample surface. Also, their fracture toughness was measured before and after ageing in Ringer's solution using the Vickers indentation technique. The set of experiments involved in this study was planned using a two-level–four-factor factorial design, and the results were analysed using the Yates algorithm. Among the main variables, the yttria content showed the greatest effect on the fracture toughness reduction of the samples studied, the same applying to the immersion temperature on what concerns the increase in the microstructure degradation of the samples after aging in Ringer's solution.

1. Introduction

Until the late 1960s, the use of permanent orthopaedic implants was limited by the lack of efficient and safe mechanisms of attachment between them and the surrounding tissue. Those implants were, almost without exception, made from metallic alloys which showed high strength and good corrosion resistance, but possessed very poor tissue adhesion and biocompatibility. As a consequence, a large number of devices needed to be removed within a relatively short time ($\approx$ 5 years) after implantation due to loosening and mechanical failure [1, 2].

One of the first attempts to solve this problem involved the use of porous ceramic coatings in order to provide long-term anchoring of the implant to the surrounding tissue [1].

Since then, the development and use of ceramic materials in biomedical applications have been continuously increasing [3–6].

Recently, several studies have been focused on the use of yttria-doped tetragonal zirconia polycrystal ceramics (Y-TZPs) as a biomaterial for the manufacturing of ceramic femoral heads and dental implants, because of their higher fracture toughness and strength when compared to alumina. However, there is still some controversy about the time-dependent deterioration of the mechanical properties of zirconia ceramics, due to phase transformation in an aqueous environment [7, 8]. Therefore, the present work aims at the study of possible time-dependent changes on the fracture toughness and microstructure of two Y-TZP ceramics with different yttria contents, when immersed in Ringer's solution.

2. Materials and methods

2.1. Methodology

The set of experiments involved in this study was planned using a two-level–four-factor factorial design, and the results were analysed using the Yates algorithm and the Student's test of significance [9].

Table I shows the variables and levels studied in the present work.

TABLE I Variables and levels studied in the present work

Variable	Description	Levels (−)	(+)
A	Sintering schedule	1450 °C/4 h	1550 °C/2 h
B	Ageing temperature	RT (30 °C)	80 °C
C	Ageing time	1 week	8 weeks
D	Yttria content (mol %)	2	3

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TABLE II Physical and chemical properties of the zirconia powders

Sample	Composition (mol %)						Average crystallite size (nm)	Surface area (m^2/g)
	ZrO_2	Y_2O_3	Na_2O	Al_2O_3	Fe_2O_3	SiO_2		
2Y	98	1.9	3.6×10^{-2}	$< 6.1 \times 10^{-3}$	3.1×10^{-3}	$< 4.1 \times 10^{-3}$	24.3	18.7
3Y	97.1	2.9	4.7×10^{-2}	$< 6.2 \times 10^{-3}$	1.6×10^{-3}	$< 4.2 \times 10^{-3}$	24.9	16.1

2.2. Materials

The zirconia powders used in the present work were provided by the Tosoh-Co., Japan. Table II shows some of the physical and chemical properties, according to the manufacturer.

2.3. Sample preparation

Sixty cylindrical specimens of 12 mm diameter and 5 mm thick were isostatically pressed at 200 MPa. The green bodies were divided into two groups, each of which was sintered according to the sintering schedule presented in Table I. The samples were then polished to a 1 μm finish, for those specimens undergoing X-ray diffraction analysis (XRD) and Vickers indentation tests, and 0.25 μm for those undergoing SEM analysis. The latter were also thermally etched and coated with a 30 nm layer of gold.

2.4. Sample characterization

The tetragonal phase content of the sample surfaces after ageing in Ringer's solution were determined by XRD analysis, using a Cuk_α radiation, with an accelerating voltage of 27.5 kV and a tube current of 12.5 mA.

The grain size of the sintered samples were obtained by SEM analysis of the polished surfaces.

The fracture toughness of the polished samples was determined by the Vickers indentation technique, employing a load of 153 N for the 2Y samples, and 98.1 N for the 3Y samples. The KI_c values were calculated using the following equation [10]:

$$KI_c = \frac{0.0319\,P}{a\,l^{0.5}}$$

where P = applied load (N) and a, l = crack lengths (m).

In order to evaluate the effect of physiological environment on the fracture toughness and microstructure degradation of Y-TZP, the samples were immersed in Ringer's solution, according to the schedule presented in Table I.

3. Results and discussion

3.1. Grain size

The grain sizes of the sintered samples are shown in Table III.

It was observed that there were no significant changes in the grain sizes of the sintered samples due to the different sintering schedules employed. All samples showed an average grain size of approximately 0.28 μm.

TABLE III Grain sizes of the sintered samples

Sample	Grain size (μm)
2Y 1450 °C/4 h	0.27 ± 0.01
2Y 1500 °C/2h	0.28 ± 0.01
3Y 1450 °C/4 h	0.28 ± 0.01
3Y 1500 °C/2 h	0.27 ± 0.01

TABLE IV Application of the Yates algorithm to the 2^4 factorial design

Exp.	%t	Effect	t_i	KI_c	Effect	t_i
(1)	100	97.85	-	20.69	14.05	-
a	100	− 0.28	1.63	20.78	0.40	1.69
b	98.0	− 4.30	25.50	17.17	− 2.33	9.80
ab	97.5	− 0.28	1.63	15.50	− 0.88	3.70
c	100	− 3.18	18.83	15.81	− 2.08	8.74
ac	100	− 0.15	0.89	17.73	0.91	3.83
bc	91.0	− 3.18	18.83	12.51	0.28	1.18
abc	89.5	− 0.15	0.89	13.36	− 0.12	0.52
d	100	1.70	10.08	11.84	− 5.30	22.28
ad	100	0.23	1.33	12.24	0.10	0.44
bd	100	1.70	10.08	11.65	1.79	7.53
abd	100	0.23	1.33	10.79	− 0.17	0.72
cd	100	0.58	3.41	9.94	1.61	6.77
acd	100	0.10	0.59	12.65	− 0.18	0.74
bcd	94.9	0.58	3.41	11.15	0	0
abcd	94.7	0.10	0.59	10.92	− 0.30	1.25

3.2. Effect of ageing on microstructure and fracture toughness degradation

In order to evaluate the significance of the results obtained through the Yates algorithm, the Student's test of significance was employed, with an α risk of 0.005 and with eight degrees of freedom, which gives a critical test statistic parameter t_v of 3.83.

Table IV shows the application of the Yates algorithm to the 2^4 factorial design used in the present work, where a = sintering schedule; b = immersion temperature; c = immersion time; d = yttria content; $\%t$ = tetragonal phase content; KI_c = fracture toughness; t_i = calculated test statistic parameter.

Based upon the results presented in Table IV, it was concluded that among the main variables, yttria content showed the greatest effect on fracture toughness

reduction of the samples studied, and immersion temperature had the most effect on increase in microstructure degradation of the samples.

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References

1. S. F. HULBERT, F. A. YOUNG, R. S. MATHEWS, J. J. KLAWITTER, C. D. TALBERT and F. H. STELLING, *J. Biomed. Mater. Res.* **4** (1970) 433.
2. L. L. HENCH, in "Biomaterials and clinical applications" (Elsevier Science Publishers B. V., Amsterdam, 1987) p. 23.
3. P. DUCHEYNE, *J. Biomed. Res.* **21** (1987) 219.
4. L. L. HENCH, *J. Amer. Ceram. Soc.* **74** (1991) 1487.
5. W. BUERGER and G. WILLMAN, in Proceedings of the 6th International Symposium on Ceramics in Medicine, Philadelphia, November, 1993, edited by P. Ducheyne and D. Christiansen. (1993) p. 299.
6. P. F. JAMES, *J. Non. Cryst. Solids* **181** (1995) 1.
7. K. SHIMIZU, K. MAKINOUCHI, P. KUMAR, T. KOTOURA, T. YAMAMURO, M. OKA and T. NAKAMURA, *J. Biomed. Mater. Res.* **27** (1993) 729.
8. I. THOMPSON and R. D. RAWLINGS *Biomaterials* **11** (1990) 505.
9. A. D. RICKMERS and H. N. TODD, in "Statistics– an introduction" (McGraw Hill, New York, 1967).
10. C. D. PONTON and R. D. RAWLINGS, *Mater. Sci. Technol* **5** (1988) 865.