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Effect of different surface treatments on the hydrothermal degradation of a 3Y-TZP ceramic for dental implants

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ABSTRACT

Objectives. Implant surface modifications are intended to enhance bone integration. The objective of this study was to assess the effect of different surface treatments on the resistance to hydrothermal degradation, hardness and elastic modulus of a 3Y-TZP ceramic used for dental implants.

Methods. Samples grouped according to their surface morphologies (AS, as-sintered; C, coated; P, dry-polished; R, roughened; PA, polished and annealed; RA, roughened and annealed) were subjected to accelerated hydrothermal degradation (LTD) by exposure to water steam (134 °C, 2 bars) for 100 h. The *t–m* phase transformation was quantified by grazing incidence X-ray diffraction (GIXDR) and by combined focused ion beam and scanning electron microscopy (FIB-SEM). Elastic modulus and hardness before- and after prolonged aging (100 h) were assessed by nanoindentation.

Results. AS and C specimens presented a better resistance to hydrothermal degradation than P and R samples. After prolonged aging, the depth of the monoclinic transformed layer ranged from 11 μm to 14 μm. Hydrothermal degradation led to a significant decrease of elastic modulus and hardness.

Significance. Surface treatments affected the resistance to hydrothermal degradation of the 3Y-TZP ceramic. Dry mechanical surface modifications should be avoided since a high *t–m* transformation rate associated to the initial monoclinic content was observed. Annealing was useful to reverse the initial *t–m* transformation, but did not improve the resistance to hydrothermal degradation.

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1. Introduction

Since the inception of modern dental implantology, titanium has been used as prime implant material due to its excellent osseointegration and favorable mechanical properties [1]. In recent years though, ceramic implants were developed as part of a strategy aimed at avoiding any intraoral metal. Such implants are made of zirconium oxide polycrystals which are stabilized in their tetragonal form by 3 mol% yttrium oxide and hence referred to as 3Y-TZP. Compared to other oxide ceramics, 3Y-TZP is considered a "high performance ceramic" due to its superior flexural strength and fracture toughness. Moreover, zirconia may be accurately processed to complex geometries using CAD–CAM [2–5] or low-pressure injection molding [6] and therefore is applicable to dental implantology [7–9].

Research on zirconia implants typically focuses on their mechanical properties, fatigue resistance and surface modifications which may enhance bone integration. As to the latter, a number of methods were developed to texture the surface and produce micro-porosity. For instance, the surfaces were roughened by sandblasting the implants with Al_2O_3 powders of various granularities [4,10–12], which some researchers complemented with hydrofluoric acid etching [11,13] or alkaline salt treatment [12]. In other works, a micro-porous structure was created by coating the implants' surface with a porous layer of zirconia [3]. More recently, a femtosecond laser technique was used to generate micro-pores or microgrooves at the surface and thus create a controlled surface texture [14].

Problematically, while it positively affects osseointegration [15–17] surface texturing also alters the implants' mechanical strength and fatigue resistance as the surface treatments might trigger the *t–m* transformation, build-up residual stresses and/or create critical-size defects [18–21].

As an additional limitation, 3Y-TZP ceramics are vulnerable to hydrothermal degradation, also known as low temperature degradation (LTD) [22–25]. A phenomenon in which, due to the presence of water, the *t–m* phase transformation is triggered at the ceramic surface. It results in a volume expansion of the grains, inducing surface roughening, micro-cracking and possibly loss of strength [23,24,26]. Hydrothermal degradation is time- and temperature dependent and does occur at 37 °C during the lifetime of the implanted device, as reported for explanted hip implants [27–30]. However, clinical hydrothermal degradation has not yet been documented for dental implants. This may be due either to differences in environmental conditions and applied forces or lack of studies addressing this particular issue. Moreover, it is well established that surface treatments such as polishing, grinding or sandblasting may affect the susceptibility of 3Y-TZP ceramics toward hydrothermal degradation [31–34].

The present study aimed at evaluating the resistance to hydrothermal degradation of a 3Y-TZP ceramic for dental implants. More specifically, the objectives were

1. to determine the influence of selected surface treatments on the kinetics of hydrothermal degradation and
2. to assess the elastic modulus and hardness of the 3Y-TZP before- and after accelerated aging.

2. Materials and methods

The material studied is a 3Y-TZP ceramic processed by hot isostatic pressing and sintering and used to produce a recently marketed implant system (Axis Biodental, Les Bois, Switzerland). Bar-shaped samples (13 mm × 5 mm × 3 mm) were provided by the manufacturer and assigned to four different groups according to their surface morphologies. Specifically, the first group contained 'as-sintered' specimens without additional treatment (AS, $n = 3$). After sintering, a 3Y-TZP coating was deposited on top of the surface (C, $n = 3$), using a proprietary method (EP 1 924 300). The 'polished' group (P, $n = 3$) was obtained after mechanical dry-polishing the 'as-sintered' surface with a diamond disk of grit size between 1 and 3 μm. The 'rough' group (R, $n = 3$) was obtained after dry-mechanical treatment of the 'as-sintered' surface, using a 20 μm grit diamond wheel. These two specific surfaces were added for a comparative purpose. Moreover, two 'polished-annealed' (PA group) and two 'rough-annealed' (RA group) specimens were obtained by annealing at 1050 °C for 15 min (Programat P500, Ivoclar-Vivadent, Schann, Liechtenstein) two polished respectively rough specimens.

Prior to proceeding to the tests, all specimens were ultrasonically cleaned in ethanol for 20 min (45–90 kHz, 320 W, Fisa Compact, FISA, France).

2.1. Surface characterization

Surface roughness was measured using an electro-mechanical profilometer (M1, Mahr, Göttingen, Germany) featuring a drive unit (PGK, Mahr) and a measuring sensor with a 40 nm z-resolution (MFW-250, Mahr). The specimens were mounted on an X–Y cross-slide table. They were scanned using a 2 μm radius diamond stylus at a force of 0.5 mN and a traveling speed of 0.5 mm s^{-1}. The profilometer generated frames of 21 line profiles with an interdistance of 140 μm. R_a was calculated using a Gaussian profile filter (ISO 11562:1996) [35] with the cut-off wavelength set at $\lambda_c = 0.8$ mm and the evaluation length at $l_n = 5.6$ mm.

Images of the surface were obtained using a scanning electron microscope (XL 20, Philips, Eindhoven, Netherlands).

2.2. Hydrothermal degradation (LTD)

The samples were aged for selected times between 1 and 100 h in an autoclave at 134 °C under 2 bars of water pressure. Tetragonal-to-monoclinic surface phase transformations consecutive to aging were measured using the grazing incidence X-ray diffraction technique (GIXRD) with a fixed incidence angle of 2° to ensure the analysis of the outermost surface layer (D500, Bruker AXS, Karlsruhe, Germany). Under these conditions, the X-ray penetration depth is 1.2 μm (as determined by the AbsorbDX software, DIFFRACplus BASIC Evaluation Package, Bruker, Karlsruhe, Germany). XRD profiles were generated in a range of 26–33° 2θ with a step size of 0.01° and a counting time of 5 s/step. The integrated intensity ratio (x_m) and the volume fraction of the monoclinic phase (f_m) were determined from the X-ray diffraction patterns using Garvie and Nicholson's [36] and Toraya et al.'s [37] equations.