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Microstructure of veneered zirconia after surface treatments: A TEM study

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ABSTRACT

Objective. Clinical studies reveal that veneer chipping is one major problem associated with zirconia based dental restorations, the underlying mechanisms being still investigated. We semi-quantitatively analyzed the effects of different surface treatments (thermal etching, 35/105 μm sandblasting and coarse bur drilling (150 μm)) on the microstructure of a zirconia veneered dental ceramic.

Methods. The relative monoclinic content on zirconia surfaces was determined using X-ray diffraction (XRD). The microstructure at the zirconia–veneer interface has thereafter been investigated using transmission electron microscopy (TEM). Selected area electron diffraction (SAED) was used to qualitatively assess the depth of the stress-induced phase transformation.

Results. Sandblasting or bur drilling significantly roughened the zirconia surface. A reverse transformation of already transformed monoclinic zirconia grains back into the tetragonal polymorph has been observed after thermal veneering treatment. In TEM, the mechanically treated samples revealed a highly damaged area of 1–3 μm from the interface. The presence of monoclinic phase in veneered zirconia samples has been observed in SAED up to depths of 4 μm (35 μm sandblasted), 11 μm (105 μm sandblasted) and 9 μm (150 μm diamond drilled) below the interface.

Significance. Regardless of the treatment protocol and produced roughness, the veneering ceramic perfectly sealed the zirconia surface. XRD showed an increased amount of monoclinic phase on the surface treated zirconia. However after thermal treatment, the monoclinic phase was re-transformed into the tetragonal polymorph. TEM/SAED analysis has found indication for a greater extend of the monoclinic transformation into the bulk zirconia compared to the treatment related defective zone depth.

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1. Introduction

Zirconia is the most widely used ceramic framework material in dentistry. Due to the metastability of the tetragonal phase at room temperature, thermal and mechanical treatments can induce a martensitic phase transformation into the monoclinic polymorph [1]. As a consequence, the properties of zirconia do not only depend on the composition and microstructure, but also on external processing parameters [2].

Zirconia is commonly processed with a veneer layer. Chipping, associated with the veneering porcelain, has been reported the main clinical problem of this type of restoration [2]. The adhesion between the two materials has been investigated by mechanical testing, and cohesive failure of the porcelain or interface delamination have been observed [3,4]. The zirconia–veneer adhesion is thereby affected by the presence of chemical bonds, mechanical interlocking, defect density at the interface, wetting properties, coefficients of thermal expansion (CTE) mismatch [5,6], and cooling stresses from the sintering process [7].

Sandblasting of dental restorations is applied either to erase the trace lines created by the CAD/CAM milling [8,9], to increase the adhesion of the luting agent by providing a high surface roughness [10], or to adjust and finish a restoration [11]. Mechanical observations revealed that sandblasting does not influence porcelain adhesion [6,12] or even enhance bond strength between zirconia and the veneer [13]. However, due to applied surface treatments, tetragonal to monoclinic transformation is triggered in the surface layer and influence the mechanical performance of zirconia. Surface roughness has been correlated with zirconia–veneer interfacial failure [3] and surface residual stresses [14].

The zirconia microstructure is a key factor in the chipping discussion. Profund research on the subsurface morphology of ground Y-TZP has been performed by Munoz-Tabarez et al. [15]. To date, little structural information is available on specific dental treatments such as sandblasting or diamond drilling and on the microstructural changes after thermal heat treatment as applied upon the veneering procedure.

The purpose of this TEM study was to evaluate the microstructure of differently treated zirconia substrates at the interface with the veneering porcelain. The extent of phase transformation was evaluated by X-Ray diffraction (XRD) and a depth analysis was performed using selected area electron diffraction (SAED).

2. Materials and methods

2.1. Zirconia/porcelain sample preparation

Plates were cut (12.5 mm × 12.5 mm × 1.25 mm) with an ISOMET low speed diamond saw (Buehler, Düsseldorf, Germany) from 3Y-TZP green bodies (DOCERAM Medical Ceramics GmbH, Dortmund, Germany) and sintered at 1430 °C for 2 h in a high temperature oven (EVA1700, LINN High Therm GmbH, Eschenfelden, Germany). The sintered plates were lapped with 1 μm alumina abrasive (PM2, Logitech, Glasgow, Scotland) with 1 μm alumina abrasive and afterwards four different surface treatments were applied ($n = 5$): control; thermal etching at 1300 °C for 1 h; sandblasting with 35 μm and 105 μm alumina particles (0.4 MPa pressure, 20 mm distance, 15 s); manual drilling with a 150 μm grit diamond bur in the X and Y directions. Three samples each group were veneered (VM9 Base Dentin, VITA, Bad Säckingen, Germany) using a common dental technician's hand-slurry technique. A three-step sintering process (Vakumat 4000 Premium T, VITA) was performed at 950, 910, and 900 °C respectively for 1 min according to the manufacturer's instructions. After lapping, the final porcelain layer thickness was approximately 0.1 mm. Two additional samples each group were subjected to the same thermal treatment used for veneering, but without the porcelain layer (simulated veneering).

2.2. Roughness analysis and X-ray diffraction

Quantitative roughness was analyzed using a confocal laser scanning microscope (TCS-SL, Leica, Bensheim, Germany) in reflectance mode. Line scans ($n = 25$ each group) were taken from the zirconia surfaces at 1000-fold magnification (HC-PL Fluotar 100, NA = 0.9). Roughness values (R_a in μm) were statistically treated using ANOVA (S–N–K test, $p < 0.05$).

X-ray diffraction was used to analyze the phase composition on the zirconia samples ($n = 2$ each group) after respective treatments. The measurements were performed with a Bruker AXS Advance D8 (Karlsruhe, Germany) X-ray diffractometer in Bragg-Brentano geometry using Cu Kα radiation ($\lambda_{K\alpha} = 1.54$ Å) and a divergence slit of 0.5°. The angular range $20° < 2\theta < 80°$ was covered with a step size of $\delta(2\theta) = 0.014°$ and 1 s step count time. Two sets of samples were analyzed, with or without the thermal treatment corresponding to the veneering process. The measurements were repeated twice on the same sample in order to account for reproducibility. An approximation suggested by Garvie and Nicholson was used to calculate the relative amount of monoclinic phase from the integrated intensities of the XRD spectra [16].

$$Xm = \frac{lm(1\,1\,1) + lm(-1\,1\,1)}{lm(1\,1\,1) + lm(-1\,1\,1) + lt(1\,1\,1)}$$

Since in the present case, only the monoclinic peak (−1 1 1) could be determined, the intensity of the peak (1 1 1) was considered as zero.

2.3. Transmission electron microscopy

Cross sections from the veneered zirconia plates ($n = 3$ each group) were prepared by cutting plates (2.5 mm × 0.5 mm) and grinding them down to a thickness of 15–25 μm. Ion milling (Gatan PIPS, Pleasanton, USA) was performed in order to obtain an electron transparent area. The samples were observed by bright-field transmission electron microscopy (CM30T/STEM, Philips, Eindhoven, the Netherlands) at 300 kV acceleration voltage.

Since the zirconia consists of polycrystalline nature, diffraction rings specific for each phase could be observed. The diffraction patterns were obtained with an SAED aperture corresponding to a selected sample area of 5.3 μm in diameter and the crystallographic phases were analyzed using JEMS