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A comparative transmission electron microscopy, energy dispersive x-ray spectroscopy and spatially resolved micropillar compression study of the yttria partially stabilised zirconia - porcelain interface in dental prosthesis

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ABSTRACT

Recent studies into the origins of failure of yttria partially stabilised zirconia–porcelain veneered prosthesis have revealed the importance of micro-to-nano scale characterisation of this interface zone. Current understanding suggests that the heat treatment, residual stresses and varying microstructure at this location may contribute to near-interface porcelain chipping. In this study the chemical, microstructural and mechanical property variation across the interfacial zone has been characterised at two differing length scales and using three independent techniques; energy dispersive X-ray spectroscopy, transmission electron microscopy and micropillar compression.
Energy dispersive X-ray spectroscopy mapping of the near-interface region revealed, for the first time, that the diffusional lengths of twelve principal elements are limited to within 2–6 μm of the interface. This study also revealed that 0.2–2 μm diameter zirconia grains had become detached from the bulk and were embedded in the near-interface porcelain.
Transmission electron microscopy analysis demonstrated the presence of nanoscale spherical features, indicative of tensile creep induced voiding, within the first 0.4–1.5 μm from the interface. Within zirconia, variations in grain size and atomistic structure were also observed within the 3 μm closest to the interface.
Micropillar compression was performed over a 100 μm range on either side of the interface at the spatial resolution of 5 μm. This revealed an increase in zirconia and porcelain loading modulus at close proximities (<5 μm) to the interface and a decrease in zirconia modulus at distances between 6 and 41 μm from this location.
The combination of the three experimental techniques has revealed intricate details of the microstructural, chemical and consequently mechanical heterogeneities in the YPSZ–porcelain interface, and demonstrated that the length scales typically associated with this behaviour are approximately ±5 μm.

1. Introduction

Yttria (Y_2O_3) partially stabilised zirconia (ZrO_2) (YPSZ) is a ceramic material which has become widely used in the manufacture of dental prostheses due to its high compressive strength, appealing aesthetics, biocompatibility and toughness [1]. In dental prosthetics, zirconia frameworks are veneered with porcelain that is applied as a slurry and then "baked". This process delivers excellent surface aesthetics, and reduces the surface hardness of the artificial tooth thereby reducing wear on the enamel of opposing natural teeth [2,3].

Despite the benefits of using porcelain as an outer coating material, clinical trials have identified the primary failure mode associated with this veneering process — near interface chipping of the porcelain [4]. The origin of chipping is speculated to be associated with the residual stresses induced in both YPSZ and porcelain at the interface during manufacture [5–7].

A large number of studies have been performed on the YPSZ–porcelain interface region in order to quantify the bonding/adhesion strength [8–11], the fracture resistance [12,13] and the overall statistics of structural integrity and reliability [14] of this region. The motivation for these

Abbreviations: YPSZ, yttria partially stabilised zirconia; SEM, scanning electron microscopy; TEM, transmission electron microscopy; EDS, energy dispersive X-ray spectroscopy; SE, Secondary Electron; BSE, Back Scattered Electron; FIB, Focused Ion Beam; MBLEM, Multi-Beam Laboratory for Engineering Microscopy.

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studies has often been to optimise the prosthesis mechanical response as a function of processing conditions (sintering temperature, etc.), hence they have primarily focused on the macro-scale behaviour of dental prosthesis as a whole.

In contrast to these macro-scale investigations, recent publications have highlighted the importance of performing high resolution (micro-to-nano scale) chemical, structural and mechanical analysis in order to improve understanding of the origins of the failure occurring at the interface. Scanning electron microscopy (SEM) analysis of the interface zone has revealed the importance of crystal structure and orientation on such failures [15,16], high resolution tomography studies have highlighted the presence of cracks in the interface zone [17,18] and microtensile bonding studies have demonstrated the importance of length scale considerations in this problem [19].

The current study aims to investigate the local micron-scale changes in chemical, microstructural and mechanical properties across the YPSZ–porcelain interface of a completed dental prosthesis using Transmission Electron Microscopy (TEM), Energy Dispersive X-ray Spectroscopy (EDS) and microcompression testing. These studies have focused on two differing length scales: ±5 μm from the interface (TEM and EDS analysis) and ±100 μm from the interface (micropillar compression and EDS analysis).

One TEM study focusing on the impact of different YPSZ surface treatments on the YPSZ–porcelain interface in disc samples has previously been performed by Grigore et al. [20]. This study revealed distinct differences in the near-interface YPSZ microstructure of these specially prepared samples and therefore that YPSZ surface preparation is highly influential in the YPSZ–porcelain bonding characteristics. In contrast to this study, in the present TEM analysis our focus is instead placed on mapping both the YPSZ and porcelain near-interface regions. Importantly, the study was carried out over much larger distances than previously considered, and on a sample representative of clinically relevant copings.

EDS analysis at the YPSZ–porcelain interface has also previously been performed in disc samples to determine the impact of different thermal processing routes on zirconium and silicon concentrations within the first few microns of the interface [21]. In the present analysis, we consider the distribution of all elements detected by EDS, and also compare the results obtained at the two differing length scales (±5 μm and ±100 μm) examined in this study.

In terms of microscale mechanical characterisation, nanoindentation has previously been performed to determine the bulk mechanical properties of both porcelain [22] and YPSZ [23,24]. A number of previous micro-compression studies on various forms of zirconia have also been reported [25,26], which revealed clearly the length scale dependence of the behaviour of this material. As no such analysis has been performed on dental porcelain or the YPSZ–porcelain interface, using this technique brings new insights into the yield strength and modulus behaviour at microscale resolution, which is known to be critical in this region.

2. Experimental

2.1. Sample preparation

The focus of this study was an incisal YPSZ–porcelain dental prosthesis which was manufactured by dental technicians for the 'Specialists Dental Group', Singapore. This tooth was selected from a series of prosthesis manufactured for patients, and was therefore as representative as possible of the samples in which porcelain chipping had previously been observed [4].

In the manufacture of the prosthesis, a YPSZ coping was machined from a Wieland Dental Zenotec Zr Bridge green body using a computer-aided manufacturing process. Following the implementation of the YPSZ recommended sintering regime by the manufacturer, IPS e.max® porcelain (by Ivoclar Vivadent) was veneered onto the surface [27]. This process involved initially applying viscous liquid porcelain to the YPSZ, before further sintering. Multiple (mm scale) layers of porcelain of differing composition were applied to produce the desired combination of bonding strength and toughness in addition to realistic aesthetic appearance.

To facilitate access to the YPSZ–porcelain interface, a 2 mm thick cross section of the prosthesis was cut using a Buehler Isomet Diamond Saw after mounting in a polymer resin. A very slow cutting speed (25 rpm on a 127 mm diameter blade) was used to reduce the impact of cutting on the sample microstructure and residual stresses. The samples were then metallographically polished using progressive grinding up to 4000 grade grit followed by diamond polishing down to 0.1 μm diamond paste, in order to reduce further the impact of the residual stresses. A careful ultrasonic cleaning process was then applied to remove any remaining surface residue or debris. The polished prosthesis slice was mounted on a SEM stub using silver paint in order to reduce the effects of charging during SEM imaging and Focused Ion Beam (FIB) machining. The Secondary Electron (SE) SEM image of the sectioned and polished sample is shown in Fig. 1. The interface region selected for further examination was chosen to be the location at which the interface was most straight in order to reduce the effects of interface curvature on the analysis.

Lamellae for TEM observation were fabricated using the software module 'AutoSlicer' [28] and the Tescan Lyra-3 XM FIB SEM at the Multi-Beam Laboratory for Engineering Microscopy (MBLEM), Oxford, UK. A 20 × 5 μm^2 area lamella containing the YPSZ–porcelain interface with an average thickness in the range of 100–150 nm, was milled out for TEM analysis. A very low magnitude polishing current (100 pA) was used to control carefully the sample thickness while minimising the impact of gallium ion implantation.

Arrays of micropillars were FIB-milled across the YPSZ–porcelain interface using a two-step milling process: coarse milling using 2.5 nA milling current followed by fine milling at 0.75 nA. The average pillar diameter and height were ~1.9 μm and ~5 μm respectively, and an aspect ratio of ~2.5 (Fig. 2) was selected to reduce the impact of pillar buckling during compression, as successfully validated in previous studies [29,30]. The pillars were separated from each other by ~10 μm and two arrays of 10 pillars each, offset by ~7 μm, were milled in order to attain the overall spatial resolution of ~5 μm in terms of the distance from the interface, as shown in Fig. 3. The relative variation in mechanical properties parallel to the interface is believed to be such that an offset of this size should not significantly influence the properties calculated. This is further demonstrated by the consistent trends observed between these two profiles as shown in Section 3.2.

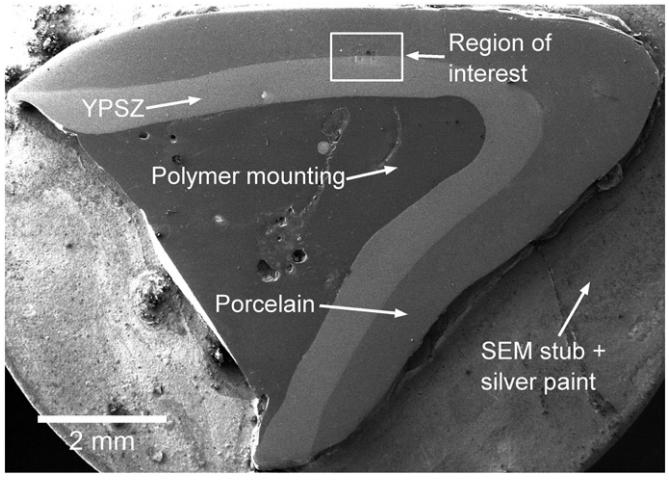

Fig. 1. SE SEM image of prosthesis cross section mounted on an SEM stub using silver paint. The outer porcelain veneer, intermediate YPSZ coping and the inner remnants of the polymer mounting (showing large voiding regions) can be observed in this image. The location of experimental analysis is also highlighted.