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Effect of tribochemical treatments and silane reactivity on resin bonding to zirconia

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ABSTRACT

Objective. The aim of the study was to assess the roughness, structure and bond strength with zirconia of four grit-blasting treatments combined with three silane types, the reactivity of which was evaluated, as well.

Methods. The grit-blasted treatments performed on zirconia (Lava) were alumina (ALU), CoJet (COJ), SilJet (SLJ) and SilJet Plus (SJP, with silica-encapsulated silane). The other two silanes selected were the S-Bond (SB, prehydrolyzed) and Clearfil Ceramic Primer Plus (CP, prehydrolyzed with 10-MDP). The activity of the silanols in the silanes was evaluated by FTIR spectroscopy. Optical profilometry and Raman microspectroscopy were used for the assessment of roughness (Sa, Sz, Sdr parameters) and structure (monoclinic volume-Vm) of zirconia, before (REF) and after grit-blasting, and a shear bond strength (SBS) with a flowable resin composite, for the investigation of the bonding capacity of the treatments.

Results. Only SB demonstrated reactive silanols. CP and the SJP silanes were mostly in a polymerized siloxane state. Roughness was increased after grit-blasting as follows: ALU > SLJ,SJP > COJ > REF (Sa,Sz) and ALU > SLJ,COJ,SJP > REF (Sdr). ALU demonstrated the highest Vm (7.52%) from all other treatments (4.16–4.81%) and the REF (0%). COJ and SLJ showed the highest SBS (14–15.94 MPa) regardless of the silane type used. SJP showed no significant differences from SLJ-SB and COJ-SB. Weibull analysis showed a reliability (β) ranking of COJ, SJP, SLJ, ALU-CP > ALU-SB > REF and a characteristic life (η) ranking of COJ, SLJ, ≥SLJ-SB, SJP, ALU ≥ ALU-SB,REF-CP > REF-SB.

Significance. The reactivity of the silanes used showed great variations to support a predictable effect in all treatments. CP with deactivated silanols demonstrated a) the most reliable and strongest treatment with a silica-rich powder (COJ), despite the lowest Sa,Sz substrate values and b) high strength with a low-silica powder (SLJ) with higher Sa,Sz substrate values. Therefore, it may be concluded that 10-MDP greatly contributes to the bonding mechanism of the silane containing primers.

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1. Introduction

Grit-blasting of dental zirconia surfaces with alumina or silica–alumina particles followed by the use of properly functionalized methacrylate coupling agents have been accepted as the most efficient and durable surface treatments for resin bonding [1,2]. The main advantages of this combined procedure are a) the chemical bonding of coupling agents with zirconia and the surface retained particles, and b) the mechanical interlocking of the luting agents or restoratives on the rough surface, following polymerization and shrinkage. This combination has been shown to overwhelm the deficiencies of using only coupling agents for chemical bonding [3,4] or grit-blasting without coupling agents [5]. Use of low pressures (<0.3 MPa) and small particle sizes (<70 μm) has been advocated to reduce the extent of microcrack formation and control the tetragonal (t) to monoclinic (m) phase transformation, at the surface and subsurface zones [6,7]. This is important, since the advantages of increased zirconia strength due to the transformation-induced residual compressive stresses after grit-blasting [8] are minimized by microcrack formation and structural destabilization [9,10].

For zirconia surfaces grit-blasted with crystalline alumina (α-Al_2O_3), phosphate functionalized methacrylate monomers have been proposed as coupling agents, which bond with alumina implanted particles and the uncoated zirconia surface [2]. For tribochemical treatments, where silica-treated alumina particles are included in the alumina powder, silane coupling agents are used, which bond with the fractions of the hydroxylated amorphous silica particles retained on the zirconia surface [2]. However, the tribochemical coating is not uniform with only small fraction of zirconia surface coverage [11]. Because of the low bonding capacity of silanes to zirconia [12], acidic ethanol solutions of silane with phosphate coupling agents have been introduced [13]. In these products hydrolyzed γ-methacryloxypropyl trimethoxysilane (MPTMS) is usually mixed with 10-methacryloyloxydecyl dihydrogen phosphate (10-MDP). MPTMS is the most commonly silane used in dental applications, since its methacrylate functionality matches that of most dental resins [14], whereas 10-MDP is a phosphate monomer capable of bonding with a wide range of substrates [12]. MPTMS has been incorporated, as well, in universal adhesives containing 10-MDP, in order to expand their applications in ceramic bonding and repair. However, it has long been stated that mixing hydrolyzed silanes with dental monomers possessing –OH groups, deactivates the silanol (Si–OH) groups via condensation reactions [14]. This has been confirmed in silane containing universal adhesives, where no Si–OH groups were traced [15], resulting in inferior bonding of these adhesives with silica-rich substrates as compared to that of separate MPTMS treatments [15,16].

Recently, a new tribochemical treatment, containing encaplulated MPTMS in silica microspheres, has been introduced for simultaneous roughening and silanization of the substrate. This treatment has been found equally efficient to traditional tribochemical methods for resin bonding to a Co–Cr alloy [17] and zirconia [18]. The status of the encapsulated silane is unknown. Based on the sealing procedure of the silane-filled silica microspheres (exposure to ambient humidity) [19], it can be assumed that siloxane polymers have been formed.

The aim of the present study was a) to evaluate the silanol reactivity of two commercially available silane primers (based on MPTMS and MPTMS/10-MDP), and a silica-encapsulated MPTMS silane, b) to assess the effects of the grit-blasting treatments on zirconia roughness and structure, and c) to investigate the bond strength of a resin composite to zirconia, as mediated by four grit-blasting treatments and the corresponding silanes. The null hypothesis was that no differences are expected in the silanol reactivity, grit-blasting effects on zirconia roughness and structure, and bond strength between the grit-blasting/silane treatments employed.

2. Materials and methods

The materials used in the study are listed in Table 1.

2.1. Silanol reactivity

A drop of each of the CP and SB silanes were applied on an infrared Ge window. The solvents were evaporated with a stream of nitrogen gas for 20 s and immediately after transmission FTIR spectra were obtained employing a spectrometer (Spectrum GX, Perkin-Elmer, Bacon, UK) under the following conditions: 4000–600 cm^{-1} wavenumber range, 4 cm^{-1} resolution and 30 scans co-addition. The presence of methoxy (2840, 1190, 1083, 850–800 cm^{-1}), siloxane (1120–900 cm^{-1}) and silanol groups (3430, 904 cm^{-1}), were considered as an indicator of the activity of freshly applied silane films. As controls served a non-hydrolyzed MPTMS silane (Dynasylan, Evonik Industries AG, Essen, Germany), an aged SB solution (SBA, 2 years at 37 °C) and a 10-MDP sample (Ivoclar Vivadent, Schaan, Liechtenstein). For SJP, which contains silane in a silica-encapsulated form, a small amount of the powder was placed on the diamond crystal of an attenuated total reflectance (ATR) accessory with ZnSe lenses (Golden Gate, Specac, Kent, UK), crashed under pressure applied with a metallic anvil and spectra were taken as before. The depth of the ATR analysis was estimated as to 2 μm at 1000 cm^{-1}. These tests were performed in triplicate.

2.2. Roughness measurements

Disk-shaped zirconia specimens (Ø = 10 mm, h = 2 mm, n = 20) were prepared and polished with SiC papers (320–1000 grit size) in a grinding-polishing machine (Dap-V, Struers, Ballerup, Demark) and ultrasonicated for 5 min, in distilled H_2O. The specimens were randomly assigned into four groups (n = 4/group) corresponding to the grit-blasting treatments with 50 μm α-Al_2O_3 (ALU), CoJet (COJ), SilJet (SLJ), SilJet Plus (SJP), and a reference polished zirconia group (REF). The central part of the specimens was blasted for 5 s with each powder using an intraoral sandblaster (Microetcher IIA, Danville Materials) operated at 2.3 bar air pressure (0.23 MPa, 0.47 L/s flow rate), 5 mm distance and 90° angle. All specimens were dried with oil-free dry air for 20 s and then examined by optical interferometric profilometry. An optical profiler (Wyko NT1100, Veeco, Tuscon, AZ, USA) was used as follows: Mirau lens, 40×

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