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Heat treatment-improved bond strength of resin cement to lithium disilicate dental glass-ceramic

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ABSTRACT

This study investigated the influence of different hydrofluoric acid (HF) concentrations and heat treatments applied to a lithium disilicate dental glass-ceramic (EMX) on surface morphology and micro-shear bond strength (μSBS) to resin cement. Five HF concentrations (1%, 2.5%, 5%, 7.5% and 10%) and four different heat treatments applied before etching were assessed: 1. etching at room temperature with no previous heat treatment (control group); 2. HF stored at 70 °C for 1 min applied to the ceramic surface at room temperature; 3. HF at room temperature applied after a hot air stream is applied perpendicularly to the ceramic surface for 1 min; 4. the combination of previously heated HF and heated EMX surface. The etching time was fixed for 20 s for all groups. Etched EMX specimens were analyzed on field-emission scanning electron microscope (FE-SEM) and the μSBS was carried out on a universal testing machine at a crosshead speed of 1 mm/min until fracture. For the control groups, FE-SEM images showed greater glassy matrix dissolution and higher μSBS for 7.5% and 10% HF concentrations. The previous heat treatments enhanced the glassy matrix dissolution more evidently for 1%, 2.5% and 5% and yielded increased μSBS values, which were not statistically different for 7.5% and 10% HF concentrations (control group). HF concentrations and previous heat treatments did show to have an influence on the etching/bonding characteristics to lithium disilicate dental glass-ceramic.

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1. Introduction

Dental ceramics have gained great notoriety in the last decade due to the remarkable improvement of their mechanical strength and ability to mimic dental tissues. Clinical indications comprise restoring tooth tissues that were decayed/fractured, replacement of unsatisfactory/failed clinical dental restorations or to re-configure the anatomical shape of mal-positioned teeth with an improvement on the design and the esthetic appearance of the smile. Nowadays, glass-ceramics are among the most commonly used indirect restorative materials in Dentistry.

Among glass-ceramics, some materials are reinforced by lithium disilicate crystals, and have recently become a popular restorative material for esthetic and functional rehabilitations [1].

The first lithium disilicate glass-ceramic introduced in the dental market was the IPS Empress II (Ivoclar Vivadent, Schaan, Liechtenstein) in 1998. This glass-ceramic contained around 60 vol% of lithium disilicate crystals dispersed in an amorphous vitreous phase and commercially available in different shades and opacities. The IPS e.max Press (Ivoclar Vivadent, Schaan, Liechtenstein) (EMX), a later version of IPS Empress II, was released in 2005 with some changes in the microstructure. EMX is a bioceramic composed by refined lithium disilicate crystals ($\pm$ 70 vol%-$Li_2Si_2O_5$. Crystals: 3–6 μm in length) embedded in a glassy matrix (information provided by the manufacturer) and it is indicated either as a full-contour restoration (monolithic) or as a core for further porcelain veneering [2]. It presents statistically similar occlusal wear as natural enamel [3] as well as translucency and high strength as a monolithic ceramic [4]. These factors, combined with the emerging demand for metal free restorations, explain the widespread use of lithium disilicate glass-ceramics [4].

The lithium disilicate glass-ceramics can be chemically bonded to tooth tissues by methacrylate-based materials, such as resin

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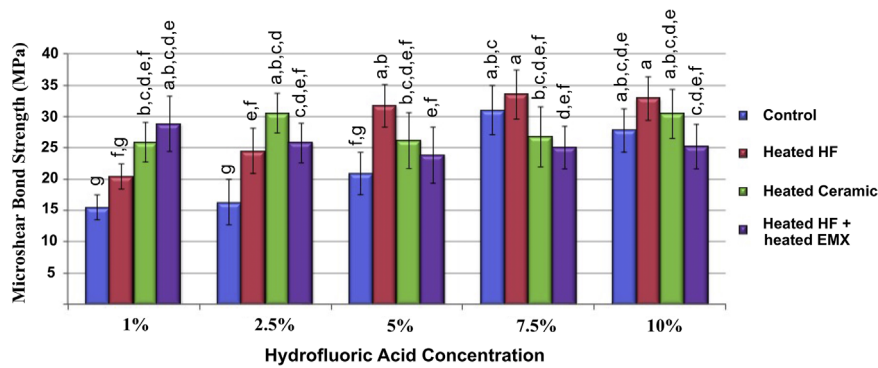E-mail address: sundfeldneto@gmail.com (D. Sundfeld).

Fig. 1. Average values of micro-shear bond strength with error bars (± Standard Deviation). Columns with different lowercase letters in vertical position indicate significant statistical difference (two-way ANOVA—Tukey's multiple comparisons test, p < 0.05).
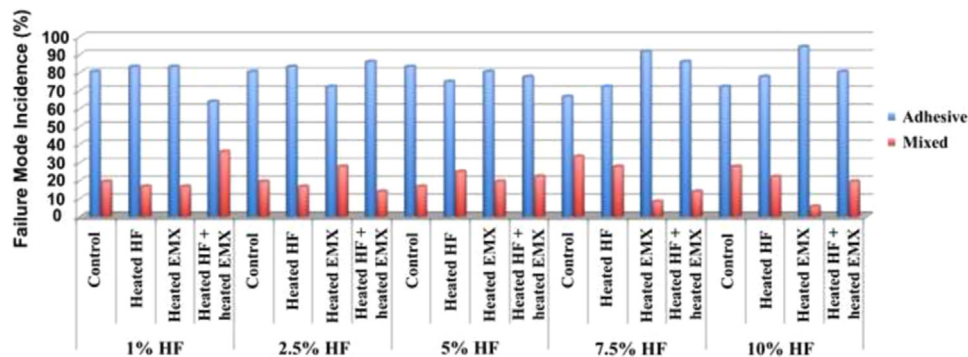

Fig. 2. Failure mode results of debonded resin cement specimens. Hydrofluoric acid (HF).

cements, and their interaction is one of the key factors to long-term clinical success [5]. Ideal bond to dental glass-ceramics is achieved by the sum of two techniques: (1) surface modification in order to increase surface area; (2) chemical bonding via silane coupling agent, which makes possible to bond an inorganic material (glassy matrix) to the resin cement (organic compound). Despite its inherent brittleness [6], resin cement bonding is able to strengthen the dental ceramic [7].

Regarding physical surface modifications of EMX, no other method has proved to be as efficient as etching with hydrofluoric acid (HF) [8–13]. HF acid etching increases the roughness [7], therefore the surface energy and wettability [14,15], and selectively dissolves the glassy matrix, exposing lithium disilicate crystals, which is essential to increase the micromechanical retention between restoration and resin cement [9,10,16]. Based on scientific and clinical evidence, HF etching followed by silane application are necessary and have become the most widely accepted surface treatments for glass-ceramics [5].

The manufacturer recommended that IPS Empress II should be etched with 10% HF for 60 s at the time it was released on the market. However, the etching time was later modified to 20 s according to the findings obtained by Spohr et al. [17]. Today, the manufacturer recommends that EMX should be etched with 4.8% HF for 20 s. However, clinically, the optimal HF acid etching time and concentration to treat glass-ceramic is not clear [7]. Since HF is hazardous to soft tissue, lower HF concentrations have been evaluated to reduce the risk of tissue damage [18]. Unfortunately, previous study has reported that HF concentrations ranging from 1% to 2.5% were not able to provide adequate bond strength to ceramic substrate [16].

In an attempt to improve the bonding potential, some researchers [19–21] have reported increased roughness/surface area/bond strength when hot etched solutions were applied onto zirconia dental ceramic. Moreover, Liu et al. [22] reported increased

bond strength to zirconia dental ceramic when 48% hydrofluoric acid was previously heated to 100 °C. To date, there are not investigations concerning the assessment of previous heat treatments on the etching morphology/interfacial bond strength of lithium disilicate glass-ceramic.

Thus, the aim of this study is to assess the effects of previous heat treatments applied to five different hydrofluoric acid concentrations and to ceramic surface on the etched surface morphology and micro-shear bond strength (μSBS) between lithium disilicate dental glass-ceramic (EMX - IPS e.max Press) and one commercial resin cement.

2. Materials and methods

2.1. IPS e.max Press blocks

Two hundred and sixty ceramic blocks of IPS e.max Press (8 mm × 8 mm × 3 mm), shade LT A2, were fabricated according to the manufacturer's instructions. The entire detailed laboratorial steps used to fabricate the specimens are reported in a previous study [16].

2.2. IPS e.max Press surface treatments

After being divested, the ceramic blocks were embedded in polyester resin (Resapol T208, Difibra/Fiberglass Ltda, Mogi das Cruzes, SP, Brazil) in rigid polyvinyl chloride (PVC) tubes and subjected to wet polishing with 1000-, 2500-and 4000-grit silicon carbide abrasive papers (Buehler, Lake Buff, IL, USA) in order to obtain a flat, polished and standardized surface. Then, all specimens were ultrasonically cleaned in distilled water for 20 min.

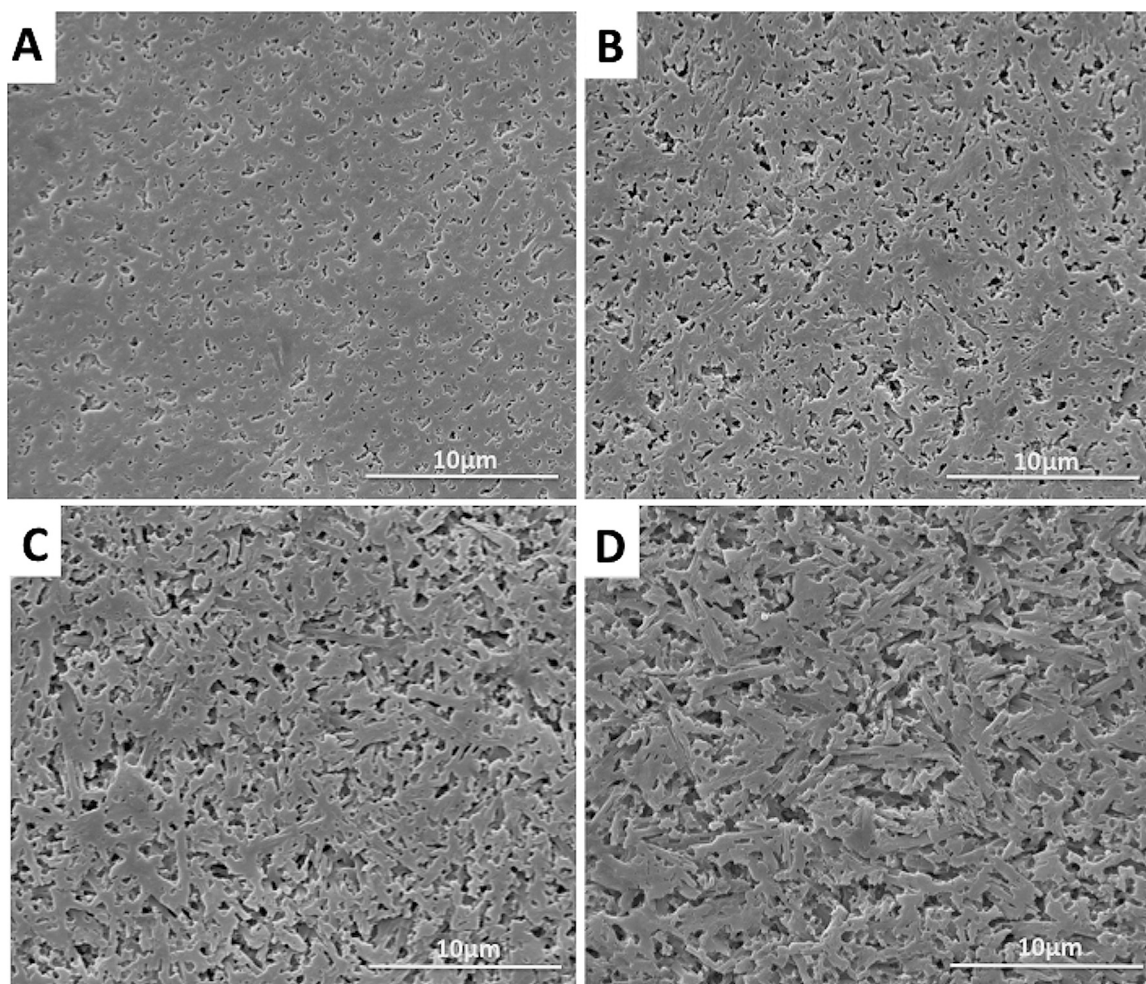The ceramic specimens were randomly assigned to 5 groups (n=52) according to the hydrofluoric acid concentrations: 1%,

Fig. 3. FE-SEM images resulting from acid etching with hydrofluoric acid (HF) 1% on IPS e.max Press for 20s. Control group (A), previous HF heat treatment (B), previous ceramic surface heat treatment (C) and the combination of previously heated HF and heated ceramic surface (D). Different etching morphologies were found with higher glassy matrix removal when the heat treatments were applied.

2.5%, 5%, 7.5% and 10% (Fórmula & Ação, São Paulo, SP, Brazil). Next, each group was randomly distributed into four subgroups according to the previous heat treatments adopted (n=13): (1) control group (no heat treatment was performed previously to etching); (2) previously heated HF; (3) previously heated ceramic surface; (4) the combination between previously heated HF and ceramic surface. These procedures are detailed below.

2.2.1. Control group

Ceramic specimens were etched for 20 s at room temperature (25 °C ± 1), then, rinsed with an oil-free air-water spray for 30 s.

2.2.2. Hydrofluoric acid heat treatment

One drop of the HF was dispensed in an eppendorf vial (1 mL) and positioned with the lid closed in the heater, which was calibrated at 70 °C ± 5 (Thermosmart, Cap-Lab, São Paulo, SP, Brazil). After 1 min, the HF was dropped on the ceramic. The etching time was 20 s, followed by rinsing with oil-free air-water spray for 30 s.

2.2.3. IPS e.max Press heat treatment

A perpendicular hot air stream was positioned 1 cm away from the ceramic surface and applied for 1 min. Next, the HF acid was dropped onto the ceramic surface and left to etch for 20 s, followed by rinsing with oil-free air-water spray for 30 s. The

temperature of 85 °C ± 1 was recorded in the ceramic surface with an infrared-thermometer (Hikari HT-450).

All the heat treatments/etching procedures were performed in a ventilated room and the operator used individual protection equipment, protection eyeglass, rubber gloves and carbon chemical mask (Half Facepiece Reusable Respirator, 6000 series, 3M ESPE, St. Paul, MN, USA). After the etching procedure, all the ceramic specimens were ultrasonically cleaned in distilled water for 20 min and air dried.

2.3. Resin cement cylinders preparation and micro-shear bond strength (μSBS)

A silane coupling agent (RelyX Ceramic Primer, 3M ESPE, St. Paul, MN, USA) was rubbed on the etched ceramic surface for 15 s and allowed to air dry for 1 min, then hot air was applied perpendicularly to the EMX surface for 1 min to accelerate the solvent evaporation. Next, a thin layer of unfilled resin (Scotchbond MP, 3M ESPE, St. Paul, MN, USA) was rubbed on the ceramic surface and light-cured for 20 s (Valo Cordless, Ultradent Inc., South Jordan, UT, USA) with an irradiance of 1000 mW/cm^2.

Elastomer molds (Oralwash L, Zhermack, Badia Polesine, Italy) with thickness of 1 mm and three cylinder-shaped orifices (2 mm distance between them) were obtained and positioned onto the ceramic surfaces, then stabilized with adhesive tape. The orifices were filled with a resin cement (Variolink II – Base paste – shade
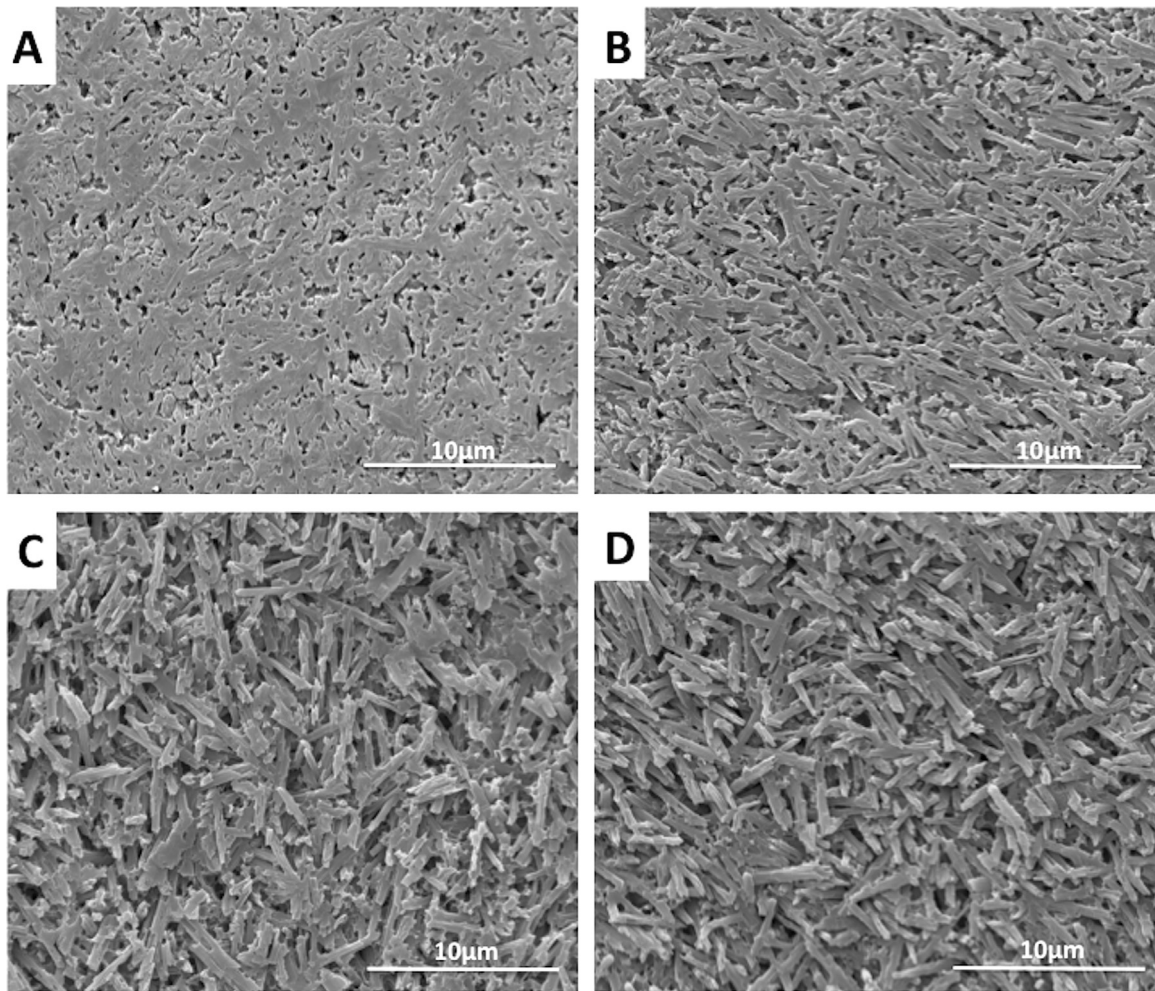
Fig. 4. FE-SEM images resulting from acid etching with hydrofluoric acid (HF) 2.5% on IPS e.max Press for 20s. Control group (A), previous HF heat treatment (B), previous ceramic surface heat treatment (C) and the combination of previously heated HF and heated ceramic surface (D). Different etching morphologies were found with higher glassy matrix removal and exposure of lithium disilicate crystals when the heat treatments were applied.

A3; Ivoclar Vivadent) and a transparent polyester strip and a glass slide were placed over the filled mold. Next, a cementation load of 250 g was applied perpendicularly to the interface for 2 min [11]. The glass slide was removed and the resin cement was light-cured for 40 s (Valo Cordless). The specimens were stored in distilled water for 24 h at 37 °C. After storage, the resin cement cylinders were checked under $40 \times$ (Olympus Corp, Tokyo, Japan) and those who presented with flaws at the interface were removed and replaced.

The PVC tubes were positioned in a universal testing machine (Model 4411, Instron, Canton, MA, USA) and an orthodontic steel wire (0.2 mm in diameter) was looped around the base of each resin cement cylinder. The micro-shear testing was calibrated with a crosshead speed of 1 mm/min until failure. Failure modes were analyzed under optical microscope (Olympus Corp, Tokyo, Japan) at $40 \times$ and classified as: adhesive (mode 1); cohesive within ceramic (mode 2); cohesive with resin cement (mode 3); and mixed (mode 4). The μSBS design is reported in a previous study [16].

2.4. Field-emission scanning electron microscopy (FE-SEM) evaluation

One random ceramic sample was selected from each group in order to characterize the etched ceramic surfaces. The ceramic blocks were mounted on coded brass stubs, sputter coated with gold-palladium for 60 s at 45 mA (Denton Vacuum Desk II, Moorestown, NJ, USA) and subjected to FE-SEM (FEI Quanta 200 Environmental Scanning Electron Microscope, Hillsboro, OR, USA) operated under 20 kV. All images are represented with a $5.000 \times$ magnification (working distance of 10 mm) with 10 μm scale bars.

2.5. Statistical analysis

For each group, 12 ceramic specimens were tested and the mean μSBS value (MPa) of the three resin cement cylinders was recorded as the bond strength of each ceramic specimen. The data were analyzed with two-way ANOVA (all HF concentrations × all heat treatments) and multiple comparisons were performed using Tukey post hoc test ($\alpha=0.05$).

3. Results

3.1. Micro-shear bond strength

The μSBS values are represented in Fig. 1. No pre-testing failures occurred. The interaction between HF concentrations × heat treatments was significant ($p=0.000$). For the control group, HF concentrations influenced the bond strength ($p=0.000$), with 7.5% and 10% resulting in higher μSBS compared to 1%, 2.5% and 5%.
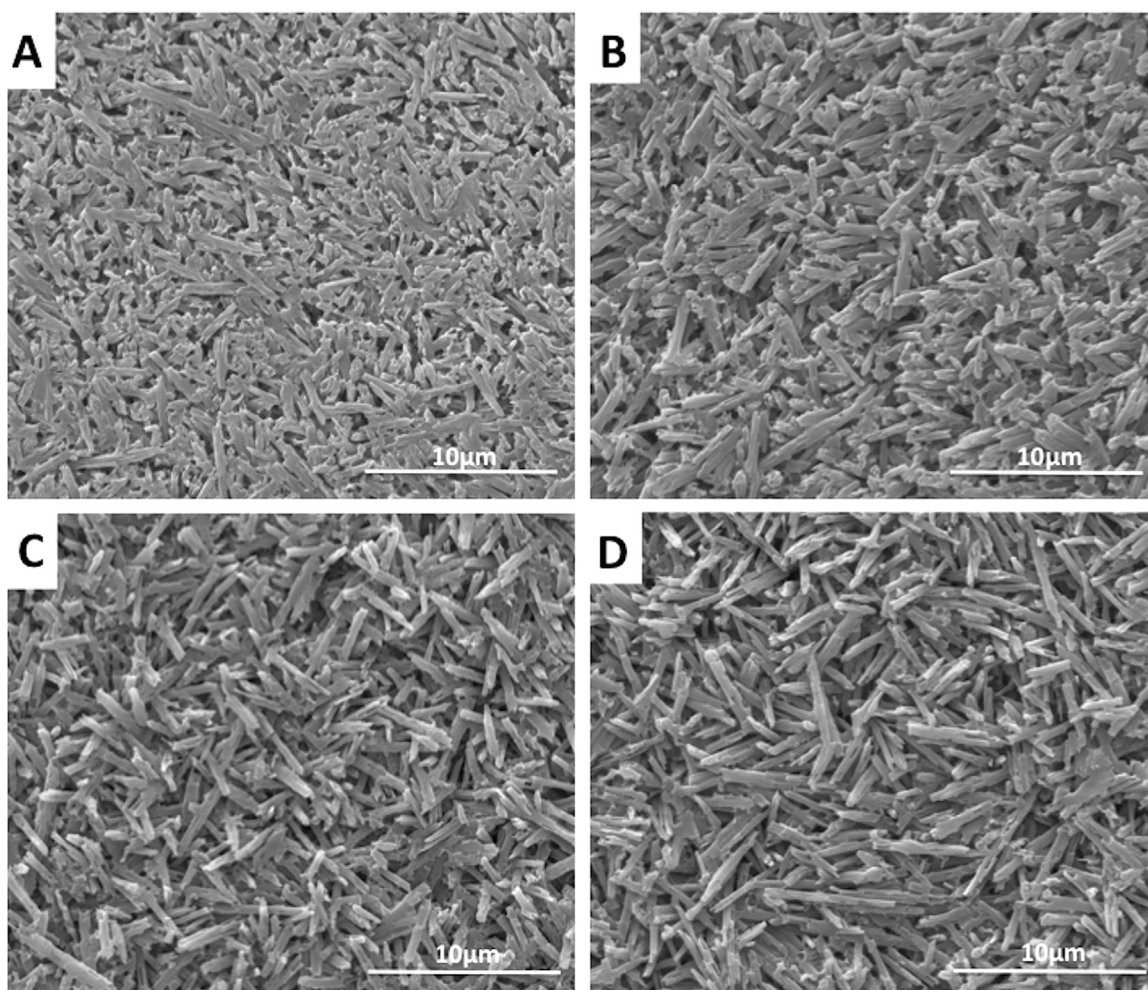
Fig. 5. FE-SEM images resulting from acid etching with hydrofluoric acid (HF) 5% on IPS e.max Press for 20 s. Control group (A), previous HF heat treatment (B), previous ceramic surface heat treatment (C) and the combination of previously heated HF and heated ceramic surface (D). Different etching morphologies were found with higher glassy matrix removal and exposure of lithium disilicate crystals when the heat treatments were applied.

Heat treatments had a statistically significant effect ($p=0.000$), with higher μSBS values presented by lower HF concentrations (1%, 2.5% and 5%).

3.2. Failure modes analysis

A descriptive analysis of the failure modes is shown in Fig. 2. The HF concentrations and the previous heat treatments have not influenced the failure modes. The predominant failure registered was adhesive (mode 1); mixed failures (mode 4) were found but with lower incidence. The heat treatments did not pre-dispose the ceramic to cohesive failure, as none was recorded. No cohesive failure within resin composite was verified.

3.3. FE-SEM images

Figs. 3, 4, 5, 6 and 7 represent, respectively, the effects of 1%, 2.5%, 5%, 7.5% and 10% HF and the resulting interaction with the heat treatments. Greater glassy matrix dissolution and exposure of lithium disilicate crystals was found when 7.5% and 10% HF concentrations were applied at room temperature (control group). The heat treatments influenced on the etching morphology but to a greater extent for HF 1%, 2.5% and 5% (Figs. 3, 4 and 5). For 7.5% and 10% HF, the heat treatments seemed to have a slight influence of the etching surface morphology.

4. Discussion

The present report evaluated the influence of heat treatment applied to HF and to ceramic surface previously to the etching step of a lithium disilicate glass-ceramic, aiming to enhance the etching effect of HF on IPS e.max Press. Through micro-shear bond strength assessment and FE-SEM images, a clear effect of temperature was confirmed.

The hydrofluoric acid is obtained by the reaction between calcium fluoride and sulfuric acid:

$$CaF_2 + H_2SO_4 \rightarrow 2HF + CaSO_4 \tag{1}$$

Thus, when applied to glass-ceramics, HF attacks the silicon-oxygen bonds (silanol) [23] based on the affinity of fluoride to silicon that is higher than to oxygen [18] and consequently is capable to selectively remove the glassy matrix (silicon – SiO_2):

$$4HF + SiO_2 \rightarrow SiF_4 + 2H_2O \tag{2}$$

In general, the initial chemical reaction rate depends on the concentration of the reactants, the temperature, pressure and on the coefficients a, b and c – $aA + bB + cC \rightarrow$ Product [23,24].

The concentration relates directly with how efficiently the HF will attack/remove the glassy matrix. The higher the initial concentration, the higher the concentration of ionized HF available to react with the glassy matrix. In contrast, heat acts as a catalyst by strongly speeding up the chemical reaction as temperature rises
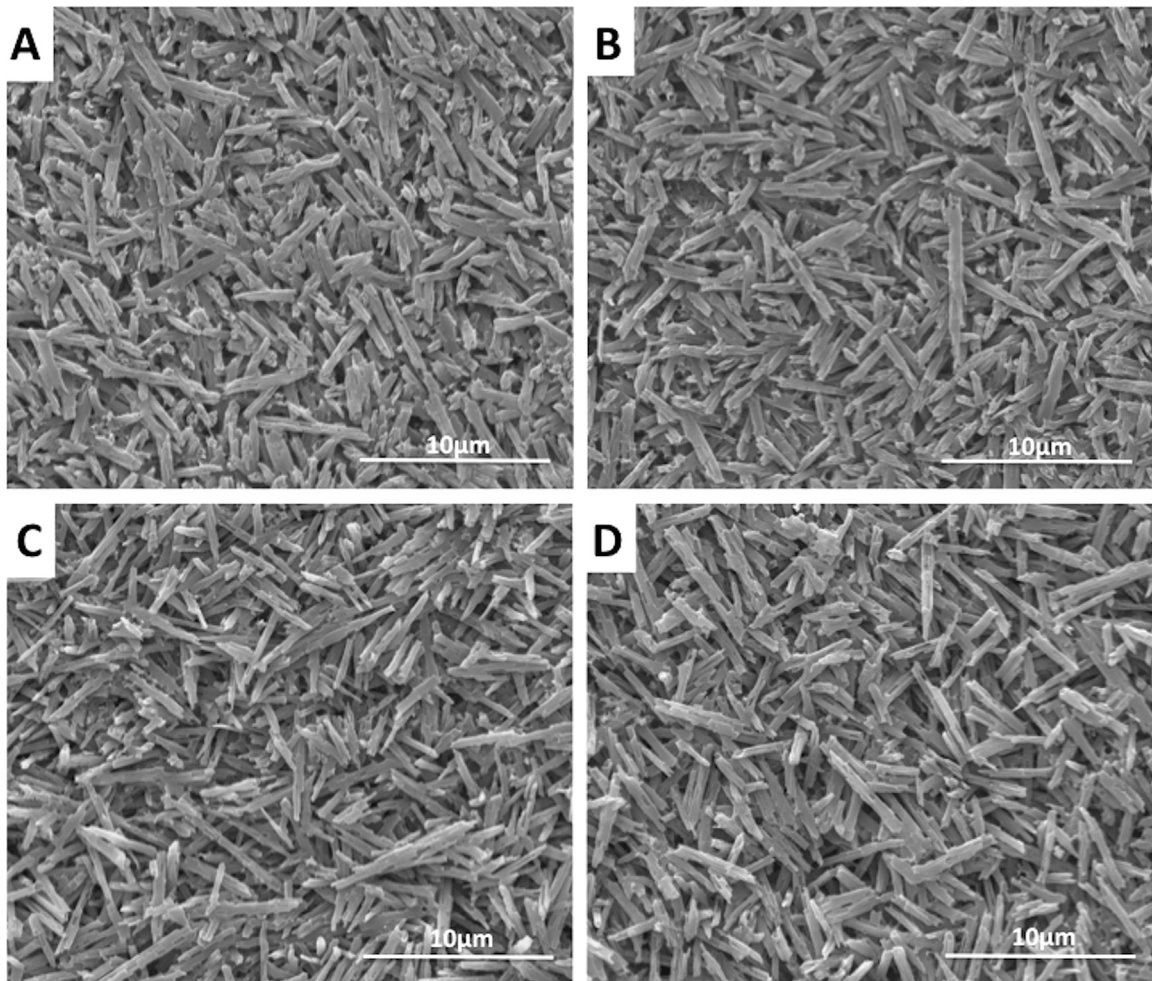
Fig. 6. FE-SEM images resulting from acid etching with hydrofluoric acid (HF) 7.5% on IPS e.max Press for 20 s. Control group (A), previous HF heat treatment (B), previous ceramic surface heat treatment (C) and the combination of previously heated HF and heated ceramic surface (D). The heat treatments had a slight effect on the glassy matrix removal.

[24], without interfering with the concentrations. This happens because the additional energy that is provided by temperature to the system due to the increased entropy generates a higher mobility of the molecules [25]. This means that there is a greater likelihood for collisions (with more energy) of ionized HF with the ceramic surface, which might increase the silanol breaking. Also, heating might have accelerated the replacement of the first oxygen by a fluoride ion, which is a slow and rate-determining reaction step [23]. The result is the formation of a greater concentration of silicon tetrafluoride (SiF_4) due to the faster consumption of the reactants (faster reaction rate) that happens at the established etching time (20 s). Therefore, an increased surface area compared to control groups is generated, explaining why pre-heating lower concentrations of acid resulted in higher glassy matrix removal and enhanced bond strength compared to control groups.

By conserving the same concentration of HF, the combination of heated 1% HF+heated ceramic surface led to a two-fold increase in μSBS compared to the control group. It is also important to note that with the association of both heat treatments, the μSBS of the 1% HF was statistically similar to 10% HF at room temperature. Lower concentrations of hydrofluoric acid are preferable due to its hazardous inherent characteristics [16,18]. However, when not previously pre-heated, it has been demonstrated that lower concentrations of HF could barely physically alter the ceramic surface, which reduced its contact to resin cement and, consequently,

resulting in lower μSBS [16]. This situation may lead to early debonding and to an unsuccessful restorative dental treatment. On the other hand, exposed lithium disilicate crystals, due to glassy matrix removal, act as retentive sites for resin cement micromechanical interlocking, resulting in higher μSBS [16,26]. Hence, as heat treatments led 1% and 2.5% HF to present similar behavior to 10% HF, adopting lower pre-heated concentrations may be considered a safer choice to etch dental glass-ceramics.

A slight decrease in μSBS was verified when the combination of heated HF + heated ceramic surface was applied for 7.5% and 10% HF concentrations. A reasonable explanation is that the increased reactiveness due to heat treatments associated with a higher concentration may have lead to higher depth of dissolution of the glassy matrix, which can be deducted due to the higher amount of "loose" lithium disilicate crystals at the surface (Figs. 6D and 7D). Those "unattached" crystals may have hampered the penetration of the bonding methacrylate materials. Also, the greater removal of glassy matrix phase may have affected the quality of the chemical bond promoted by the silane, as less silica would be available at the surface, and the bond between ceramic-resin material may have been impaired.

The higher frequency of adhesive failures (mode 1) suggest that: (1) the μSBS truly assessed the interfacial bond strength as even higher values still presented a higher frequency of adhesive failures; (2) the heat treatments have not weakened the EMX as no cohesive failures (mode 2) were verified after the μSBS.
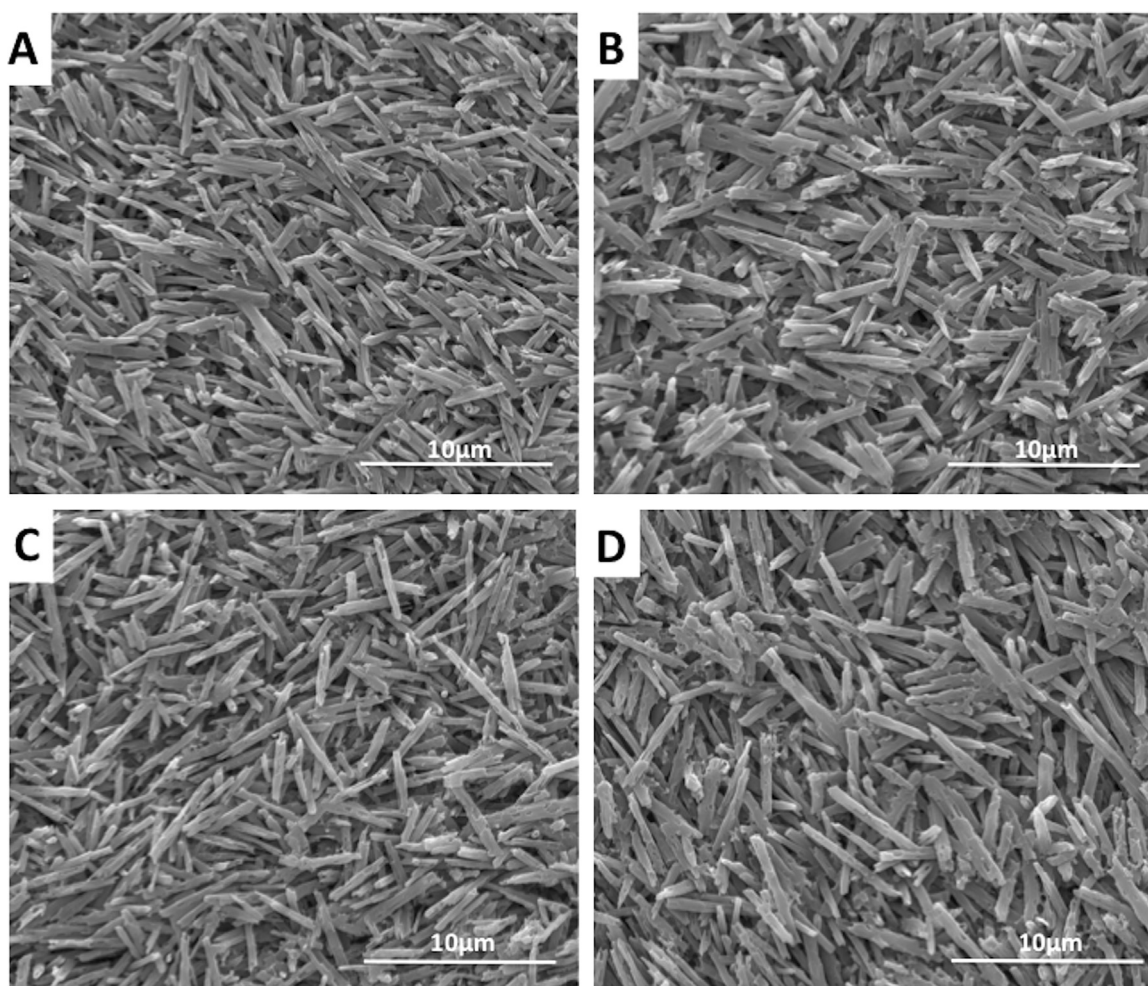
Fig. 7. FE-SEM images resulting from acid etching with hydrofluoric acid (HF) 10% on IPS e.max Press for 20 s. Control group (A), previous HF heat treatment (B), previous ceramic surface heat treatment (C) and the combination of previously heated HF and heated ceramic surface (D). The heat treatments had a slight effect on the glassy matrix removal.

It is known that hydrofluoric acid is a hazardous etchant [18], especially for dental technicians and/or prosthetic dental professionals who are often exposed to HF during the etching step. Also, it can not be neglected that, despite the fact that the ceramic piece is etched outside of the oral cavity, patients could be susceptible to accidental exposures/spills when hydrofluoric acid is applied on the surface of broken ceramic restorations for further bonding of a methacrylate-based restorative material in case of intraoral repairs. Thus, a noteworthy criticism on the safety/biological advantage of the heat treatments may arise from the results obtained in this study: with the increased reactivity of HF, even for lower acid concentrations, preheating the ceramic/acid may be considered as harmful as higher concentrations of acid. Although preheated HF 1% groups showed similar μSBS values to HF 10% groups, heated HF 1% groups did show a shallower etching pattern compared to HF 10% groups. Therefore, we hypothesize, based on the etching pattern, that lower preheated HF concentrations would potentially be less harmful than higher ones. Further studies are necessary to investigate specifically the biological effects of this approach.

The present study proposed the heating treatments as a way to increase the HF reactiveness to etch a lithium disilicate glass-ceramic. The main objective of this investigation was successfully accomplished because it is clear that heat treatment does have an influence on the etched surface morphology and bond strength to lithium disilicate glass-ceramic. Other variables, such as different temperatures and etching times, are worth investigating further in combination.

5. Conclusions

It has been shown that hydrofluoric acid can physically modify the lithium disilicate glass-ceramic surface according to its concentration. With higher HF concentrations (7.5% and 10%), a greater glassy matrix removal and exposure of lithium disilicate crystals are observed. This increased surface area results correlated with higher bond strength to resin cement.

The present study also reports a technique that is capable of increasing the etching potential of hydrofluoric acid concentrations by performing a heat treatment on the hydrofluoric acid and/or on the ceramic surface previously to the etching step itself. By those approaches, it was possible to enhance the etching effect (especially for HF 1%, 2.5% and 5%) and the bond strength values.

Therefore, it may be suggested to clinically use previously heated 1% or 2.5% hydrofluoric acid instead of higher concentrations when etching to lithium disilicate glass-ceramic.

Acknowledgments

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