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Kiara Serafini Dapieve

INFLUÊNCIA DE TRATAMENTOS DE SUPERFÍCIE E CIMENTOS RESINOSOS NO COMPORTAMENTO MECÂNICO E ADESIVO DE CERÂMICAS VÍTREAS

Santa Maria, RS
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Tese apresentada ao Curso de Doutorado do Programa de Pós-Graduação em Ciências Odontológicas, da Universidade Federal de Santa Maria (UFSM), como requisito parcial para a obtenção do grau de Doutora em Ciências Odontológicas – **Ênfase em Prótese Dentária.**

Orientador: Prof. Dr. Luiz Felipe Valandro
Coorientador: Prof. Dr. Cornelis Johannes Kleverlaan

Santa Maria, RS
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Kiara Serafini Dapieve

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Aprovado em 10 de março de 2023:

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Santa Maria, RS
2023

Com amor,

aos meus pais Valmor e Graciele

e ao meu irmão Guilherme.

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(Jean de la Bruyere)

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Se a alma não é pequena.
Quem quer passar além do Bojador
Tem que passar além da dor.
Deus ao mar o perigo e o abismo deu,
Mas nele é que espelhou o céu.
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(Isaac Newton)

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(Mario Quintana)

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Onde o conhecimento está apenas num homem, a monarquia se impõe. Onde está num grupo de homens, deve fazer lugar à aristocracia. E quando todos têm acesso às luzes do saber, então vem o tempo da democracia.
(Victor Hugo)

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(Carl Gustav Jung)

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(Cora Coralina)

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All my life, though some have changed
Some forever, not for better
Some have gone and some remain
All these places had their moments
With lovers and friends, I still can recall
Some are dead, and some are living
In my life, I've loved them all.
(The Beatles)

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De tudo ficaram três coisas...

A certeza de que estamos começando

A certeza de que é preciso continuar

A certeza de que podemos ser interrompidos antes de terminar

Façamos da interrupção um caminho novo

Da queda, um passo de dança

Do medo, uma escada

Do sonho, uma ponte

Da procura, um encontro.

(Fernando Sabino)

RESUMO

INFLUÊNCIA DE TRATAMENTOS DE SUPERFÍCIE E CIMENTOS RESINOSOS NO COMPORTAMENTO MECÂNICO E ADESIVO DE CERÂMICAS VÍTREAS

AUTORA: Kiara Serafini Dapieve
ORIENTADOR: Luiz Felipe Valandro
COORIENTADOR: Cornelis Johannes Kleverlaan

Esta tese é composta por cinco estudos. O estudo 1 avaliou a influência de tratamentos da superfície de cimentação da cerâmica (apenas aplicação de agente de união à base de silano – AU, ácido fluorídrico – HF ou HF+AU) e de regimes de armazenamento (com ou sem envelhecimento) no comportamento à fadiga de restaurações simplificadas de dissilicato de lítio (DL). Foi observado que o envelhecimento influenciou negativamente o desempenho de todos os grupos. O microembricamento mecânico resultante do condicionamento ácido preponderou no comportamento em fadiga e, assim, o AU foi dispensável para o desfecho avaliado. O estudo 2 avaliou a influência de tratamentos da superfície de cimentação da cerâmica (HF ou primer cerâmico autocondicionante - PC) e regimes de armazenamento (com ou sem envelhecimento) na resistência de união de um cimento resinoso com duas viscosidades (alta ou baixa) à cerâmica de DL através de um ensaio de microcisalhamento. Além disso, investigou a viscosidade dinâmica dos componentes do sistema de cimentação. Diferenças no conteúdo de carga inorgânica afetaram a viscosidade dos cimentos resinosos, que por sua vez influenciou a resistência de união a uma cerâmica de DL, de acordo com cada tratamento de superfície e regime de armazenamento. O estudo 3 avaliou a influência da viscosidade de um cimento resinoso (alta ou baixa) e do modo de carregamento (estático ou fadiga cíclica) na resistência de união ao cisalhamento do cimento resinoso a substratos de DL e dentina. O cisalhamento sob fadiga cíclica apresentou efeitos deletérios no comportamento adesivo e nas probabilidades de sobrevivência de conjuntos cimentados de DL, independentemente da viscosidade do cimento resinoso. Em contraste, a viscosidade do cimento resinoso afetou a resistência de união e as taxas de sobrevivência do substrato dentinário submetido ao modo de carregamento cíclico, no qual uma baixa viscosidade resultou em melhor desempenho. O estudo 4 avaliou o efeito da viscosidade de um cimento resinoso (alta ou baixa) e de tratamentos da superfície de cimentação da cerâmica (HF+AU ou PC) de coroas usinadas de DL no comportamento mecânico à fadiga. HF+AU/alta viscosidade e PC/baixa viscosidade apresentaram o melhor desempenho em fadiga. O comportamento das coroas cimentadas foi dependente das alterações topográficas da superfície cerâmica e na capacidade do agente cimentante em preencher as irregularidades. O estudo 5 avaliou o efeito de tratamentos da superfície de cimentação da cerâmica (HF+AU ou PC), da viscosidade de um cimento resinoso (alta ou baixa) e de regimes de armazenamento (com ou sem envelhecimento) no desempenho à fadiga de restaurações simplificadas de DL e de cerâmica feldspática (FEL). O envelhecimento pode influenciar negativamente o comportamento mecânico das restaurações vítreas simplificadas. Além disso, as "relações de microestrutura cerâmica - condicionamento da superfície cerâmica - viscosidade do cimento resinoso" modularam a performance em fadiga das restaurações simplificadas de DL e FEL.

Palavras-chave: Cerâmica vítrea. Fadiga. Resistência de união. Reologia. Tratamentos de superfície.

ABSTRACT

INFLUENCE OF SURFACE TREATMENTS AND RESIN CEMENTS ON THE MECHANICAL AND ADHESIVE BEHAVIOR OF GLASS-CERAMICS

AUTHOR: Kiara Serafini Dapieve
PROMOTER: Luiz Felipe Valandro
CO-PROMOTER: Cornelis Johannes Kleverlaan

This thesis is composed of five studies. Study 1 evaluated the influence of ceramic bonding surface treatments (application of silane-based coupling agent only – CA, hydrofluoric acid – HF or HF+CA) and storage regimes (with or without aging) on the fatigue behavior of simplified lithium disilicate (LD) restorations. It was observed that aging negatively influenced the performance of all groups. The micromechanical interlocking resulting from the acid etching prevailed in the fatigue behavior; thus, the CA was dispensable for the evaluated outcome. Study 2 evaluated the influence of ceramic bonding surface treatments (HF or self-etching ceramic primer – E&P) and storage regimes (with or without aging) on the bond strength of a resin cement with two viscosities (high or low) to LD ceramic through a microshear test. Furthermore, it investigated the dynamic viscosity of the components of the luting system. Differences in inorganic filler content affected the viscosity of resin cements, which in turn influenced the bond strength to a LD ceramic, according to each surface treatment and storage regime. Study 3 evaluated the influence of the resin cement viscosity (high or low) and the loading mode (static or cyclic fatigue) on the shear bond strength of the resin cement to LD and dentin substrates. Shear under cyclic fatigue presented deleterious effects on the adhesive behavior and survival probabilities of LD bonded sets, regardless of resin cement viscosity. In contrast, resin cement viscosity affected bond strength and survival rates of dentin substrate subjected to cyclic loading mode, in which a low viscosity resulted in better performance. Study 4 evaluated the effect of resin cement viscosity (high or low) and ceramic bonding surface treatments (HF+CA or E&P) of machined LD crowns on mechanical fatigue behavior. HF+CA/high viscosity and E&P/low viscosity showed the best fatigue performance. The behavior of bonded crowns was dependent on the topographic alterations of the ceramic surface and on the luting agent's ability to fill in the irregularities. Study 5 evaluated the effect of ceramic bonding surface treatments (HF+CA or E&P), resin cement viscosity (high or low), and storage regimes (with or without aging) on the fatigue performance of simplified LD and feldspathic ceramic (FEL) restorations. Aging can negatively influence the mechanical behavior of simplified glass restorations. Furthermore, the "ceramic microstructure relationships - ceramic surface conditioning - resin cement viscosity" modulated the fatigue performance of LD and FEL simplified restorations.

Keywords: Bond strength. Fatigue. Glass-ceramic. Rheology. Surface treatments.

SUMÁRIO

1 INTRODUÇÃO GERAL

A pluralidade de sistemas cerâmicos desenvolvidos para a tecnologia de desenho e manufatura assistida por computador (*Computer-aided design/Computer-aided manufacturing*, CAD/CAM) está em constante evolução para atender à crescente demanda por restaurações estéticas, biocompatíveis e resistentes (SPITZNAGEL; BOLDT; GIERTHMUEHLEN, 2018; BACCHI; CESAR, 2022). Em um contexto restaurador, as cerâmicas vítreas, tais como a feldspática e o dissilicato de lítio, se destacam pela capacidade óptica em mimetizar os tecidos dentários e pela versatilidade clínica (SILVA et al., 2017; SPITZNAGEL; BOLDT; GIERTHMUEHLEN, 2018).

Indubitavelmente, as tecnologias para o processamento e para a fabricação de restaurações impulsionam melhorias para aumentar a previsibilidade de tratamentos dentários confeccionados com materiais cerâmicos vítreos (BACCHI; CESAR, 2022). Por outro lado, a usinagem e o condicionamento ácido, procedimentos inerentes a esse contexto, podem introduzir defeitos no material, tornando a superfície interna de cimentação da cerâmica uma zona crítica e sensível a defeitos (ALVES et al., 2022). Dessa forma, esses procedimentos podem afetar negativamente as propriedades mecânicas e de superfície (MAY; FRAGA; MAY, 2021), sendo esse efeito deletério motivado pela natureza friável das cerâmicas odontológicas (ALVES et al., 2022).

Nesse sentido, uma união estável e duradoura entre restauração, agente cimentante e substrato é necessária para minimizar os efeitos deletérios dos defeitos introduzidos na superfície cerâmica (KELLY; BENETTI, 2011; TIAN et al., 2014). Considerando o conteúdo cristalino e a microestrutura das cerâmicas vítreas, a cimentação adesiva apresenta-se como uma estratégia padrão-ouro para esse fim (MANSO et al., 2011; TIAN et al., 2014; MATINLINNA et al., 2018; DA ROSA et al., 2022). Ademais, quando a restauração cerâmica é submetida a cargas oclusais, a cimentação adesiva propicia uma melhor distribuição de tensões e, consequentemente, aumenta a resistência à fratura do conjunto restaurador por um efeito de fortalecimento (MAY et al., 2012; LIM; LEE, 2017; DA ROSA et al., 2022).

Entre os princípios-chave de uma adesão duradoura e estável está a introdução de uma superfície rugosa e ávida ao microembricamento interfacial, de uma molhabilidade de superfície e de uma adaptação adequada para interação íntima entre os componentes do conjunto restaurador (MARSHALL et al., 2010). Assim, previamente à cimentação adesiva, tratamentos de superfície químicos e mecânicos têm sido propostos para aprimorar a adesão dos agentes cimentantes às cerâmicas vítreas (MANSO et al., 2011; TIAN et al., 2014;

SCHERER et al., 2018; DAPIEVE et al., 2020; LIMA et al., 2022). Esses mecanismos podem ser alcançados, em um primeiro momento, através do condicionamento com ácido fluorídrico, que remove seletivamente a matriz vítrea da superfície cerâmica, aumenta a rugosidade e a área de adesão, facilitando a penetração do cimento resinoso e a formação de um microembricamento mecânico com o agente de cimentação (BRENTEL et al., 2007; MENG; YOSHIDA; GU, 2010; PROCHNOW et al., 2017; SCHERER et al., 2018; DAPIEVE et al., 2020).

Após o condicionamento ácido, recomenda-se a aplicação de um agente de união à base de silano, uma molécula bifuncional que tem como objetivo conectar quimicamente o cimento resinoso e a cerâmica (MATINLINNA et al., 2018). No entanto, a degradação na cavidade oral de agentes de união contendo silano ainda gera questionamentos pela instabilidade da molécula e pela tendência de hidrólise – quebra da ligação na presença de água (BRENTEL et al., 2007; MATINLINNA et al., 2018; DIMITRIADI et al., 2019). De fato, os mecanismos sinérgicos de retenção micromecânica e adesão química suscitam dúvidas sobre a contribuição de cada tratamento (condicionamento ácido vs. agente união à base de silano) (DIMITRIADI et al., 2019). Estudos *in vitro* com restaurações de dissilicato de lítio adesivamente cimentadas submetidas a protocolos de envelhecimento mostraram uma alta taxa de descimentação (55%) (SCHERER et al., 2018) e, também, um decréscimo considerável na carga de falha para fadiga (39%) (DAPIEVE et al., 2020), quando a superfície cerâmica foi tratada apenas com agentes de união à base de silano (sem condicionamento ácido prévio). Logo, essas evidências sugerem que o embricamento micromecânico resultante da aplicação do ácido fluorídrico parece ser um fator preponderante no desempenho a longo prazo dessas restaurações.

Uma alternativa ao protocolo convencional de tratamento de superfície com ácido fluorídrico seguido da aplicação de agente de união à base de silano é a utilização de um primer cerâmico autocondicionante (SCHERER et al., 2018; SCHESTATSKY et al., 2019; TRIBST et al., 2019; DAPIEVE et al., 2020) que apresenta como vantagens a simplificação da técnica (passo único) e a menor toxicidade em relação ao ácido fluorídrico. Relatos clínicos de curto tempo de acompanhamento (2 anos) demonstram resultados promissores, como a ausência de trincas ou descimentação de restaurações feldspáticas (laminados e coroas) adesivamente cimentadas (SOUZA et al., 2020; NASCIMENTO et al., 2021). Estudos *in vitro* indicam que a performance em fadiga de cerâmicas vítreas tratadas com esse primer é semelhante e, em alguns casos, superior ao tratamento convencional (SCHESTATSKY et al., 2019; TRIBST et al., 2019; DAPIEVE et al., 2020). Esse aprimoramento no comportamento sugestiona que o tratamento com o primer reduz o número de defeitos na superfície cerâmica devido às alterações

topográficas mais suaves do que as produzidas pelo condicionamento com ácido fluorídrico (TRIBST et al., 2019; DAPIEVE et al., 2020).

Considerando que tratamentos de superfície podem produzir alterações topográficas e que o material cerâmico apresenta baixa tolerância a defeitos, a presença e o desenvolvimento de falhas superficiais têm um papel dominante no acúmulo de danos, na redução do tempo de vida útil e no comportamento à fadiga de materiais cerâmicos (KELLY et al., 2017). Em vista disso, o agente cimentante deve ser capaz de infiltrar e selar os defeitos superficiais, reduzindo a população de defeitos e aumentando a energia necessária para a falha catastrófica das restaurações cerâmicas (NAVES et al., 2010; MAY et al., 2012; SPAZZIN et al., 2016; SPAZZIN et al., 2017; VENTURINI et al., 2018; BARBON et al., 2019; DAPIEVE et al., 2020). Contudo, o agente cimentante nem sempre é capaz de penetrar em todas as irregularidades da superfície cerâmica. Essa infiltração deficiente pode ocorrer devido a diferenças na viscosidade do cimento resinoso, que pode afetar a capacidade do agente cimentante em fluir adequadamente sobre os defeitos da superfície (SPAZZIN et al., 2017; BARBON et al., 2019).

Nessa perspectiva, é importante compreender que dentre as características de um cimento resinoso, destaca-se a viscosidade, que determina a medida da resistência de um líquido às forças que tendem fazer o material fluir (SILIKAS; WATTS, 1999; AL-AHDAL; SILIKAS; WATTS, 2014). A viscosidade pode ser modificada pelo pré-aquecimento de um agente resinoso (MARCONDES et al., 2020; BARBON et al., 2022), alterando a proporção da matriz resinosa (LEE; UM; LEE, 2006; BARBON et al., 2019), usando diferentes composições (LEE; UM; LEE, 2006; BEUN et al., 2009; COELHO et al., 2019) ou por diferentes tamanhos e morfologias das cargas inorgânicas (LEE; UM; LEE, 2006). Dessa forma, cimentos resinosos preenchidos com mais carga tendem a ser mais viscosos e podem diferir de materiais mais fluídos no que diz respeito ao potencial de preencher defeitos em padrões resultantes de usinagem dos sistemas CAD/CAM e de protocolos de condicionamento de superfície (ácido fluorídrico seguido da aplicação de agente de união à base de silano ou primer cerâmico autocondicionate) (SPAZZIN et al., 2017; SCHERER et al., 2018; SCHESTATSKY et al., 2019; DAPIEVE et al., 2020).

O mecanismo de reforço promovido pela cimentação adesiva sugere a criação de uma camada híbrida entre cimento resinoso – cerâmica e torna a presença de porosidades ou falhas não preenchidas potenciais defeitos críticos, que podem concentrar tensões e reduzir a resistência do conjunto (ADDISON, SODHI, FLEMING, 2010; SPAZZIN et al., 2017; DAPIEVE et al., 2020; DA ROSA et al., 2022). Assim, compreende-se que os efeitos da

viscosidade e que a capacidade do cimento resinoso de preencher completamente os defeitos introduzidos na topografia da cerâmica podem influenciar no desempenho adesivo e mecânico de sistemas restauradores.

É indispensável ressaltar que o objetivo final de uma “adesão ideal” é produzir uma interface resistente e durável (MARSHALL et al., 2010). Dessa forma, compreender os componentes das interfaces que são submetidos aos tratamentos de superfície, cimentados adesivamente e ainda sujeitos aos desafios em função dos efeitos da água e da ciclagem térmica são essenciais para melhorar a previsibilidade das ligações que compõem o sistema restaurador (MARSHALL et al., 2010). Mesmo que metodologias clássicas de aferição da resistência de união tenham o importante papel para avaliar diferentes variáveis a fim de aprimorar os sistemas restauradores, Van Meerbeek e Frankenberger (2020) entendem que a resistência adesiva estática tradicionalmente executada não reflete a carga dinâmica que as interfaces são submetidas na cavidade oral, ou seja, a união entre “cerâmica – agente cimentante – substrato” deveria ser sujeita a carregamento cíclico intermitente, portanto, submetida a efeitos de fadiga cíclica.

De fato, o conjunto restaurador está inserido em um ambiente oral complexo e exposto às forças de cisalhamento, tração, compressão e flexão, além da ação da temperatura e umidade ao longo do tempo (TIAN et al., 2014). Sabe-se ainda que as restaurações são continuamente carregadas e submetidas à fadiga, uma abordagem clinicamente relevante que induz uma resposta do material frente a desafios cíclicos (KELLY et al., 2017; VELHO et al., 2022). A fadiga cíclica envolve um fenômeno sinérgico de degradação em consequência de ciclos/cargas intermitentes e do crescimento lento e subcrítico de trincas, que é fortemente influenciado pelo ambiente úmido. Como resultado, esses mecanismos podem degradar e enfraquecer a estrutura do material cerâmico (KELLY et al., 2017). Os mesmos princípios podem ser válidos para a interface adesiva, de modo de que a presença de bolhas e espaços não-preenchidos expostos à aplicação cíclica de carga podem evoluir para a degradação da interface ou para a falha coesiva do cimento resinoso. Nessa condição, os materiais e as interfaces adesivas inerentes ao sistema podem enfraquecer e degradar, suscitando valores de resistência de união abaixo dos obtidos em ensaios estáticos.

Levando em consideração a alta instabilidade do agente de união à base de silano em meio aquoso, a importância de compreender a influência de tratamentos de superfície de cerâmicas e de investigar os efeitos da viscosidade de um cimento resinoso frente ao microcisalhamento, cisalhamento em fadiga, fadiga mecânica e após o envelhecimento de

conjuntos restauradores de cerâmicas vítreas adesivamente cimentadas, a presente tese foi formatada em cinco artigos científicos:

ARTIGO 1: "*Is the application of a silane-based coupling agent necessary to stabilize the fatigue performance of bonded simplified lithium disilicate restorations?"* que teve como objetivo avaliar a influência dos tratamentos da superfície de cimentação da cerâmica (somente ácido fluorídrico, agente de união à base de silano, ou ácido fluorídrico seguido da aplicação de agente de união à base de silano) e regime de armazenamento (com ou sem envelhecimento) na performance em fadiga de restaurações simplificadas de dissilicato de lítio;

ARTIGO 2: "*Adhesion to lithium disilicate glass-ceramics after aging: Resin viscosity and ceramic surface treatment effects"* que teve como objetivo avaliar a influência dos tratamentos da superfície de cimentação da cerâmica (ácido fluorídrico seguido da aplicação de agente de união à base de silano ou primer cerâmico autocondicionante), viscosidades de um cimento resinoso (alta ou baixa) e regimes de armazenamento (com ou sem envelhecimento) na resistência de união do cimento resinoso a uma cerâmica de dissilicato de lítio. Além disso, investigar a viscosidade dinâmica dos componentes do sistema de cimentação;

ARTIGO 3: "*Cyclic fatigue vs static loading for shear bond strength test of lithium disilicate and dentin substrates: A comparison of resin cement viscosities"* que teve como objetivo avaliar influência da viscosidade de um cimento resinoso (alta ou baixa) e diferentes modos de carregamento (resistência ao cisalhamento estático ou resistência ao cisalhamento por fadiga), além de taxas de sobrevivência em uma cerâmica de dissilicato de lítio, considerando diferentes substratos (dissilicato de lítio ou dentina);

ARTIGO 4: "*Do resin cement viscosity and ceramic surface etching influence the fatigue performance of bonded lithium disilicate glass-ceramic crowns?"* que teve como objetivo avaliar a influência da viscosidade de um cimento resinoso (alta ou baixa) e tratamentos da superfície de cimentação da cerâmica (ácido fluorídrico seguido da aplicação de agente de união à base de silano ou primer cerâmico autocondicionante) na performance em fadiga de coroas usinadas de dissilicato de lítio;

ARTIGO 5: *"Ceramic surface conditioning, resin cement viscosity, and aging relationships affect the load-bearing capacity under fatigue of bonded glass-ceramics"* que teve como objetivo avaliar a influência de tratamentos da superfície de cimentação da cerâmica (ácido fluorídrico seguido da aplicação de agente de união à base de silano ou primer cerâmico autocondicionante), viscosidades de um cimento resinoso (alta ou baixa) e regimes de armazenamento (com ou sem envelhecimento) no desempenho à fadiga de restaurações simplificadas de cerâmicas vítreas (dissilicato de lítio e feldspática) adesivamente cimentadas.

2 ARTIGO 1: Is the application of a silane-based coupling agent necessary to stabilize the fatigue performance of bonded simplified lithium disilicate restorations?

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Is the application of a silane-based coupling agent necessary to stabilize the fatigue performance of bonded simplified lithium disilicate restorations?

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Running title: Effect of silane application on fatigue performance

Abstract

This study evaluated the influence of ceramic surface conditioning and storage regimen (baseline vs. aging) on the fatigue performance of simplified lithium disilicate glass-ceramic restorations. A total of 90 ceramic discs (Ø= 10 mm; thickness= 1.0 mm) were allocated into 6 groups (n= 15), considering 2 factors: "ceramic surface treatment" – CA (only silane-based coupling agent, Monobond N), HF (5% hydrofluoric acid etching), or HF+CA (5% HF acid etching plus silane-based coupling agent); and "storage regimen" – baseline (24 hours – 5 days of distilled water at 37ºC), or long-term aging (180 days of distilled water at 37ºC + 25,000 thermal cycles). After intaglio ceramic conditioning, adhesive bonding (Multilink N) was performed onto epoxy resin discs (Ø= 10 mm; thickness= 2.5 mm) and the bonded sets were subjected to step-stress fatigue tests (initial load: 200 N; step-size: 50 N; 10,000 cycles per step; 20 Hz). Fatigue data were analyzed using Kaplan-Meier and Weibull statistical analyses. Fractography and topography analyses were also conducted. The fatigue findings demonstrated that the performance among groups for both baseline and aging conditions maintained a tendency: the CA groups had the worst behavior (baseline: 893 N/143,667 cycles; aging: 639 N/84,179 cycles), while the surface etching with HF (baseline: 1247 N/214,333 cycles; aging: 816.67 N/128,333 cycles) and HF+CA groups (baseline: 1290 N/222,333 cycles; aging: 900 N/145,000 cycles) did not present statistical differences between them. The aging protocol reduced the performance of all groups. The groups with better fatigue performance (HF and HF+CA) did not have statistical differences regarding structural reliability (Weibull modulus). Most failures were radial cracks from the cementation interface, except for CA aging specimens, with 27% failing from debonding. The HF etching led to noteworthy surface topographical alterations. Micromechanical interlocking resulting from HF acid etching remained prevalent in the fatigue behavior. Thus, the silane-based coupling agent (Monobond N) does not need to be applied after HF etching in terms of fatigue behavior outcomes.

Keywords: Acid Etching; Cementation; Ceramics; Mechanical Phenomena; Silanes.

Highlights

- Hydrofluoric acid etching is a must for proper fatigue behavior of lithium disilicate restorations.
- Silane application alone induces a lower fatigue behavior and high rate of debonding;
- Silane application does not influence the fatigue behavior and can be disconsidered for fatigue outcomes.
- Long-term-aging induces a statistical decrease in fatigue outcomes.

1. Introduction

A stable and long-lasting bond between the substrate and the intaglio surface of a ceramic restoration is needed in order to achieve long-term clinical success (Manso et al., 2011; Matinlinna et al., 2018). In this context, luting agents are used to seal the interface between the tooth and the restoration, and the adhesive strategy shows an improvement in the mechanical properties of the assembly when compared to adhesive and non-adhesive cementation, requiring higher loads until fracture (Attia et al., 2006). Adhesive cementation provides better load distribution and acts as a strengthening mechanism of the ceramics (May et al., 2012). Since adhesive cementation provides better restoration performance, the bonding between the restoration and the substrate is also related to the intaglio surface treatment of the ceramic material (Blatz et al., 2003), and therefore the ceramic microstructure must also be considered (Addison et al., 2007).

The adhesive cementation procedure for glass-ceramics (such as lithium disilicate) involves a bonding process in the cementation surface: hydrofluoric (HF) acid etching and coupling agent application. Acid etching is responsible for selectively removing the glass matrix from the ceramic surface, creating microporosities which increase the bonding area and facilitate infiltration of the coupling agent and resin cement, thereby enabling the formation of mechanical interlocking (Brentel et al., 2007; Meng et al., 2011; Prochnow et al., 2017; Scherer et al., 2018; Dapieve et al., 2020). The acid treatment also promotes surface cleaning, increasing the surface energy by removing the low energy contaminants (Yoshida et al., 2015; Moreno et al., 2019) and enhances the density of hydroxyl groups in the ceramic surface, which is important for siloxane linkages (Matinlinna et al., 2018).

It is widely recommended to apply a silane-based coupling agent after the etching procedure, which is a bifunctional molecule derived from silicon and carbon. Its main goal for glass-ceramic silanization is to increase the chemical bonding capacity of resin cements (Dimitriadi et al., 2018). First, as a result there is the establishment of strong siloxane linkages (-Si-O-Si) through hydrogen bond formation (Brentel et al., 2007; Matinlinna et al., 2018). Second, there is a resin matrix and silane bond formation through light activation or chemical curing. As a result, resin and ceramic are chemically connected by the silane-based coupling agent (Matinlinna et al., 2018). In addition, the ceramic's surface energy is increased and the contact angle between the ceramic and fluids (such as resin cement and adhesives) is decreased as a consequence of the previous surface etching, providing the needed wettability for the silane-based coupling agent and resin cement to infiltrate into irregularities, enhancing the mechanical interlocking (Prochnow et al., 2018[b]).

However, bond degradation over time in the oral cavity is still the major concern regarding treatment with silane-based coupling agents, because this molecule is very unstable and presents an inherent tendency to hydrolyze in the presence of water (Brentel et al., 2007; Dimitriadi et al., 2018; Matinlinna et al., 2018). Also, the synergistic bonding mechanisms (micromechanical retention and chemical adhesion) have raised questions about the contribution extent of each treatment (acid etching x silane-based coupling agent) to the overall interfacial strength (Dimitriadi et al., 2018). Furthermore,

in vitro studies with adhesively bonded lithium disilicate restorations have shown that ceramic restorations presented a high debonding rate (55%) (Scherer et al., 2018) and a high drop in fatigue failure load (38.9%) (Dapieve et al., 2020) when their surface was only treated with silane-based coupling agents (without acid etching), demonstrating that micromechanical interlocking is a preponderant factor in the long-term performance of the bonded restorations.

Taking into account the high instability of silane-based coupling agent in aqueous media and the absence of studies which assess aggressive aging conditions in fatigue behavior of bonded sets, this study aimed to answer the following question: What is the influence of silane-based coupling agent surface treatment on load-bearing capacity under fatigue of simplified lithium disilicate restorations submitted to long-term aging? Therefore, this study evaluated the influence of the ceramic intaglio surface treatment (silane only vs. HF vs. HF + silane-based coupling agent) and storage regimen (baseline vs. long-term aging) on the load-bearing capacity under fatigue of simplified lithium disilicate glass-ceramic restorations. The null hypothesis tested was that the ceramic intaglio surface treatments would perform similarly for fatigue performance (before and after aging).

2. Material and methods

The composition, manufacturers, and batch numbers of the materials used in this study are described in Table 1.

2.1. Study design

The study was designed in 6 groups (n= 15), considering 2 factors (Table 2):

(i) "Ceramic surface treatment" in 3 levels: CA – only silane-based coupling agent application; HF – only 5% hydrofluoric (HF) acid etching; or HF+CA – 5% HF acid etching followed by silane-based coupling agent application.

(ii) "Storage regimen" in 2 levels: baseline – 24 hours up to 5 days of storage (distilled water, 37 ºC); or long-term aging – 180 days of storage (distilled water, 37 ºC) plus 25,000 thermal cycles.

Simplified assemblies were used as a sample unit. They consisted of lithium disilicate discs (Ø= 10 mm; thickness= 1.0 mm) with simulated Computer-aided design/Computer-aided manufacturing system (CAD/CAM) topography (Dapieve et al., 2020; Pilecco et al., 2021) adhesively cemented onto a substrate disc (Ø= 10 mm; thickness= 2.5 mm) (Chen et al., 2014).

2.2 Specimen preparation

Lithium disilicate glass-ceramic CAD/CAM blocks (IPS e.max CAD) were shaped into a cylinder format (Ø= 10 mm) using a coarse diamond grinding disc (Color Green, Grit of 240 μm, Buehler, Lake Bluff, USA) in a polishing machine (Ecomet/Automet 250, Buehler) under constant water-cooling. The cylinders were cut (Isomet 1000, Buehler) under water-cooling, resulting in a total of 90 ceramic discs. Both surfaces of the discs were polished (#400, #600, and #1200 SiC papers) using

manual pressure in a polishing machine (EcoMet/AutoMet 250, Buehler) to standardize the surfaces and achieve a thickness of 1.0 mm. Then, the cementation surface was subjected to an in-lab simulation of CAD/CAM milling topography by manually grinding the ceramic surface with light digital pressure for 15 seconds on each axis (*x* and *y*) onto a standardized size (100 mm × 50 mm) of #60 grit humidified SiC paper for each disc by a single trained operator (KSD) (Dapieve et al., 2020; Pilecco et al., 2021). The ceramic discs were subsequently crystallized according to the manufacturer's instructions (840°C, 7 minutes vacuum, Vacumat 6000 MP, VITA Zahnfabrik, Bad Säckingen, Germany). Next, the cementation surface roughness was measured on a contact surface roughness tester (Mitutoyo SJ-410, Mitutoyo Corporation, Kawasaki, Japan) taking three measurements on each axis (*x* and *y*). The average of Ra and Rz parameters (μm) of each specimen was calculated to guarantee the in-lab simulation of CAD/CAM milling topography and to standardize roughness among the specimens (mean ± standard deviation: Ra= 1.659 ± 0.136 and Rz= 10.426 ± 0.792).

As a substrate, epoxy resin discs were cut from the rod bar in a cutting machine (Isomet 1000, Buehler) into 90 discs (Ø= 10 mm). They were manually polished on both sides with wet #1200-grit SiC paper to standardize the surface and achieve 2.5 mm in thickness (Chen et al., 2014; Scherer et al., 2018; Dapieve et al., 2020; Pilecco et al., 2021).

2.3 Surface treatments and bonding procedures

The ceramic discs and epoxy resin discs were cleaned in an ultrasonic bath (1440 D, Odontobras, Ind. and Com. Equip. Med. Odonto. LTDA, Ribeirão Preto, Brazil) for 5 minutes with 78% isopropyl alcohol and distilled water, respectively, and air-dried before the bonding procedure.

The ceramic discs were randomly distributed in the following surface treatments according to experimental design (Table 2) and manufacturer recommendations:

(i) CA treatment: a silane-based coupling agent (Monobond N) was actively applied over the bonding surface for 15 seconds and kept untouched for 45 seconds.

(ii) HF treatment: the bonding surface was etched with 5% HF acid (IPS Ceramic Etching Gel) for 20 seconds, rinsed with air-water spray for 30 seconds, and air-dried. Then, the specimens were cleaned (ultrasonic bath, distilled water, 5 minutes).

(iii) HF+CA treatment: as treated by HF followed by CA procedure, aforementioned.

The bonding surface of the epoxy resin discs was etched with 10% HF acid (Condac Porcelana 10%, FGM, Joinville, Brazil) (Kelly et al., 2010; Prochnow et al., 2018[a]; Scherer et al., 2018; Dapieve et al., 2020; Pilecco et al., 2021) for 60 seconds, rinsed for 30 seconds with air-water spray, and air-dried. The discs were subsequently cleaned (ultrasonic bath, distilled water, 5 minutes). Multilink N Primer A and B (Ivoclar Vivadent) were mixed in a 1:1 ratio and applied for 30 seconds actively and air-dried to obtain a thin and uniform layer. Then, the resin cement (Multilink N, Ivoclar Vivadent) was manipulated and applied onto the ceramic disc bonding surface. Next, each ceramic disc was placed over the corresponding substrate pair under a constant load (2.5 N) on the top of the assembly. The resin

cement excess was removed with a micro-applicator and the assemblies were light-cured (Radii-cal LED curing light, SDI, Bayswater, Australia) for five exposures of 20 seconds around the assembly (0º, 90º, 180º, 270º and on the top) (Prochnow et al., 2018a; Scherer et al., 2018; Dapieve et al., 2020).

2.4 Storage regimens

The bonded assemblies from each treatment were randomly distributed into two following storage conditions (Table 2):

(i) Baseline (not aged): storage in distilled water at 37 ºC for 24 hours until 5 days before testing;

(ii) Aging (long-term): 25,000 thermo cycles (Nova Ética, São Paulo, Brazil) of 30-second baths at 5 and 55ºC with a transfer time of 5 seconds and storage in distilled water at 37ºC for 180 days before testing (Armstrong et al., 2017; Van Meerbeek and Frankenberger, 2020).

2.5 Step-stress fatigue testing

The assemblies (n= 15) were tested using the step-stress test method in an electric machine (Instron ElectroPuls E3000, Instron, Norwood, USA). Cyclic loading was applied with a hemispheric stainless-steel piston (Ø= 40 mm) at a 20 Hz frequency (Velho et al., 2020) under distilled water. An adhesive tape (110 µm) was placed on the top surface of the sample to prevent contact damage between the piston and the top surface of the assembly (Kelly, 1999). The load was initially applied with 200 N for 5,000 cycles to adjust the relationship between the specimen and the piston. Then, incremental steps of 50 N for 10,000 cycles were applied until sample failure (radial cracks or debonding). The presence of cracks was verified through light oblique transillumination (Dibner and Kelly, 2016) at the end of the 10,000 cycles of each step. The data of load and the number of cycles for failure were recorded for statistical analyses.

2.6 Fractographic analysis

All the specimens were inspected by stereomicroscope (Discovery V20, Carl Zeiss, Gottingen, Germany) after the fatigue testing, and representative samples (n= 1) were selected from each group, in which the ceramic fragments were detached with the aid of a lever instrument perpendicular to the radial crack to expose the fractured surface and access the origin of the defects. The fragments were then ultrasonically cleaned (distilled water, 5 minutes), air-dried, gold-sputtered (Edwards S150B, BOC Edwards, Burgess Hill, United Kingdom), and analyzed under Scanning Electron Microscopy (SEM, Evo LS15, Carl Zeiss, Gottingen, Germany) at 200× magnification to determine the fracture characteristics.

2.7 Topographic analysis

Additional ceramic specimens (n= 1) for each surface treatment were produced to be inspected regarding the topographical changes, microstructure features, and alterations after the surface treatment. The samples were produced and treated according to the procedures mentioned in Section 2.3. Then the specimens were gold-sputtered (Edwards S150B) and analyzed by SEM (Evo LS15) at 20,000× magnification.

2.8 Data analysis

A statistical software program (IBM SPSS Software; IBM, Armonk, USA) was used to perform the data analysis with a significance level of 0.05. Fatigue data (fatigue failure load and the number of cycles for failure) were subjected to survival analysis (Kaplan Meier and Log- Rank Mantel-cox post-hoc tests) to access the mean, confidence interval, and survival probability through the testing steps. Additionally, fatigue data were submitted to Weibull analysis (Super SMITH Weibull 4.0k-32, Wes Fulton, Torrance, United States) under the maximum-likelihood method to obtain the Weibull modulus of each group and its respective 95% confidence intervals. Fractographic and topographic analyses were qualitatively evaluated.

3. Results

Fatigue failure load, number of cycles for failure, and survival probability demonstrated that the performance among groups for both the baseline and aging conditions maintained a tendency: CA groups presented the worst behavior (baseline: 893 N/143,667 cycles; aging: 639 N/84,179 cycles), and the surface etching with HF (baseline: 1247 N/214,333 cycles; aging: 817 N/128,333 cycles) and HF+CA groups (baseline: 1290 N/222,333 cycles; aging: 900 N/145,000 cycles) did not present statistical differences between them. It is also evident that the aging procedures were able to reduce the performance of all groups, with a decrease in load and cycles in a range from 28% to 41% (Table 3, Fig. 1). For the Weibull analysis, it is highlighted that the groups with better fatigue performance (HF and HF+CA) in both aging conditions did not present statistical differences regarding mechanical structural reliability (Weibull modulus). Furthermore, intragroup reliability did not change after aging (Table 4).

One pre-test failure (debonding) was verified after aging procedures in the CA aging group and was excluded from the statistical analysis. Furthermore, the failure mode differed among the groups: the failure outcome for the HF and HF+CA (both baseline and aging) and CA baseline groups was exclusively radial crack, while 4 specimens (27%) in the CA aging group showed debonding between ceramic and substrate during the test, which was recorded (load and cycles in the step which debonding occurred) and computed in the statistical analysis (Table 3). The fractographic analysis of specimens with radial cracks showed that the failures originated from the cementation surface and that the cracks propagated perpendicularly to the compression side, where the compression curl layer is clearly visible (Fig. 2).

The topographic analysis demonstrated the potential of the surface treatments to promote the surface alterations (glassy matrix removal and pull out of lithium crystals). The CA treatment (non-etched ceramic surface) presented a smooth and regular surface, while HF treatment induced more porous and irregular surfaces as a result of the HF etching; HF+CA with a more homogeneous surface compared to HF, which demonstrated that the silane-based coupling agent layer was able to fill, at least superficially, the defects created by the etching (Fig. 3).

4. Discussion

The synergistic bonding mechanisms by micromechanical retention promoted by acid etching and chemical adhesion by the silane-based coupling agent are endorsed as a classic treatment for glass-ceramic surfaces. However, the present study shows that the reliability and fatigue performance of simplified lithium disilicate restorations both before and after a long-term aging protocol have no statistical differences when etching is performed and regardless of silane-based coupling agent application. Besides that, the coupling agent groups presented the worst mechanical behavior. Thus, the hypothesis that the surface treatments would behave similarly among them (before and after aging) was rejected.

As already known, hydrofluoric acid etching selectively dissolves the glassy matrix or pullout of the crystalline components of glass-ceramics (such as lithium crystals) and produces an irregular porous surface (Prochnow et al., 2018[a]; Scherer et al., 2018; Dapieve et al., 2020; Pilecco et al., 2021) (Fig. 3). Porous surfaces increase the surface area and thus the penetration of resins into microretentive spaces formed on the etched ceramic surface. However, there is a hypothesis that hydrofluoric acid etching can also produce insoluble silica-fluoride salts as residues or deposits on the surface, and prevent the silane-based coupling agent and/or luting resin to fully penetrate the porous ceramic surface created by etching (Maruo et al., 2017). From this viewpoint, the production of by-products with the etching procedure may explain the reason for the similarity among the groups treated with HF (with or without silane-based coupling agent), but this hypothesis still needs to be investigated in future researches.

As already mentioned, a silane-based coupling agent promotes a bond between resin composites and silica-based or silica-coated indirect restorative materials; however, the main problem stated in the literature is the weakening of the bond (degradation) in the wet oral environment over time (Brentel et al., 2007; Dimitriadi et al., 2018; Matinlinna et al., 2018). As a result, it is observed that the intaglio surface treatment with just acid etching increases bond strength to a level which diminishes the silane-based coupling agent contribution, providing bond strength values similar to silane-free treatments (Maruo et al., 2017; Dimitriadi et al., 2019). The viscosity and surface tension of the resin cement create a micro-mechanical retention pattern capable of neutralizing the chemical action of the silane-based coupling agent (Dimitriadi et al., 2019); this is in line with our fatigue findings, in that the silane-based coupling agent effect was not only negligible after long-term aging, but also before aging (Table 3 and Fig. 1). In summary, the results demonstrate that micromechanical interlocking is preponderant in the

long-term performance of simplified glass-ceramics bonded restorations (Scherer et al., 2018; Dapieve et al., 2020).

In this manner, the consequences on the topography found after surface treatments can be visualized in micrographs (Fig. 3). While the unetched ceramic surface presented a smoother surface, the acid-etched surface showed evident dissolution of the vitreous matrix and creation of retentive micro-grooves; conversely, the application of silane-based coupling agent after acid etching seems to have formed a layer, reducing the microporosities produced by etching, which agrees with the study by Moreno et al. (2019). The ceramic surface is activated when acid etched, increasing the density of the hydroxyl groups and the surface energy. This high energy in the solid tends to form bonds with other atoms which are close to the surface to reach the lower energy state. However, the energy balance after silane-based coupling agent application is modified because the chemical bonds (Si – O) on the surface and reduce the ceramic surface energy (Moreno et al., 2019). Therefore, it could be that this reaction neutralizes the main benefits of the chemical bonding of the silane-based coupling agent on the restorative assembly.

When silane-free treatments are evaluated as control groups, a comparison with silane-based coupling agent treatment to polished ceramic surfaces before and after acid-etching could provide a way to estimate the contribution of chemical bonding, because micromechanical retention was minimal due to the small roughness values. This fact can be observed in a study that showed the superiority of the treatment with silane-based coupling agent compared to silane-free treatment, although it showed very low bond strength values of lithium disilicate ceramic surfaces (3.2-6.4 MPa, depending on the silane-based coupling agent used, Dimitriadi et al., 2019). Also, the chemical bonding capacity of silane was higher in products with silanol monomers rather than products with silanol-siloxane adducts and siloxane polymers (Dimitriadi et al., 2019). This reinforcement was quite strong to balance the reduced chemical bonding capacity of several products with reduced or minimal silanol activity, which may explain our results since the silane-based coupling agent used in this study (Monobond N, Ivoclar Vivadent) did not present silanol monomers according to Dimitriadi et al. (2019). Thus, the findings of our study are restricted to the silane-based coupling agent used, and the need to evaluate coupling agents with other compositions is reinforced.

Conversely to our fatigue findings, studies evaluating bond strength still show the classic treatment with “hydrofluoric acid etching + silane-based coupling agent” as the best performance compared to treatment with hydrofluoric acid alone (Baratto et al., 2015; Lise et al. 2015; Cardenas et al., 2017; Li et al., 2019). These studies attributed this to the combined effect of micromechanical interlocking and formation of siloxane bonds (chemical bonding). However, we can emphasize that the effect of the silanization technique is dependent on the silane-based coupling agent and resin cement used. The different trends reported in the literature may be related to several factors such as silane-based coupling agents with different compositions, and therefore potentially different reactivity and stability; in addition, different silane-based coupling agent/adhesive/resin cement combinations and also different

silane-based coupling agent application methods (such as hot air temperature). In conclusion, the silanization protocol affects the bond strength of resin cement/ceramic differently, depending on the particular interaction and compositions of materials used.

It can be observed through the structural reliability (Weibull modulus), a parameter that describes the spread of load values in an asymmetrical distribution, was similar between the groups with "silane" and "acid etching + silane" treatment (Table 4) and a good Weibull fit is indicative of a single, dominant flaw type and confirmation of adequate care in testing procedures. However, it is prudent to verify the fracture initiation cause to certify this, and by the standards for Weibull analyzes in required cases (Quinn and Quinn, 2010), because it involves systematic observation and pattern recognition in failed parts which can provide information regarding failure origin and loading conditions (Bonfante and Coelho, 2016). Our fractographic images show that the failures in all groups (CA, HF, and HF+CA) originated from defects presents on the cement-ceramic interface as consequences of the specimen preparation (including the simulation of CAD/CAM topography), surface defects produced by acid etching, or even bubbles in the cement-ceramic interface (Bonfante and Coelho, 2016; Prochnow et al., 2018a; Dapieve et al., 2020; Pilecco et al., 2021).

Lastly, the aging protocol (water storage and thermocycling) was able to effectively degrade the restorative set, with a relevant decrease for all groups in fatigue failure load (28-34%) and cycles (35-41%), however without affecting the structural reliability (Table 3, Table 4). The survival probabilities (Fig. 1) clearly show the impact of long-term aging on mechanical behavior, and it is important to note that 27% of specimens in the silane-based coupling agent aging group showed debonding during the test. In fact, a combination of two mechanisms of bond weakening, i.e., hydrolysis and fatigue degradation leads to a significant decrease in bond strength that affects the overall mechanical behavior of the restorative set (Heikkinen et al., 2013). The interfacial phase of the bonded layer exposed to water over a long period of time tends to deteriorate because of the hydrolysis of the polysiloxane network between the ceramic and the polymerized intermediate resin matrix (Brentel et al., 2007; Heikkinen et al., 2013; Dimitriadi et al., 2018; Matinlinna et al., 2018; Ramakrishnaiah et al., 2018). On the other hand, thermocycling can cause repeated thermal expansion and contraction of the materials, leaching, and swelling of the components, and, then, formation of microcracks and partial disintegration of the resin matrix through repeated sorption/desorption cycles, which likely cause fatigue at the interphase (Drummond, 2008; Heikkinen et al., 2013, Ramakrishnaiah et al., 2018).

The fatigue performance of the restorative set involves a myriad of factors, such as the mechanical properties and microstructure of the ceramic and substrate, rheological characteristics of the luting agent, and finally the interaction among surface treatments and consequently the wettability of the resin cement. We only explored one brand of each material and a defined mechanical test setup in this *in vitro* study, and there are inherent limitations to these factors. Therefore, possibly suppressing the application of a silane-based coupling agent, especially the silane tested herein, which would

simplify the bonding technique and generate fewer costs, must be carefully evaluated and explored with other compositions and interactions.

5. Conclusion

- The micromechanical interlocking resulted from hydrofluoric acid etching of the intaglio surface remained as a preponderant role for higher fatigue behavior of simplified lithium disilicate restorations;
- The application of silane-based coupling agent (especially the Monobond N) after hydrofluoric acid etching can be disconsidered when taking into account the fatigue outcome observed herein.
- The application of silane-based coupling agent alone induced lower load-bearing capacity under fatigue of the bonded samples.
- Long-term aging had a detrimental effect on the fatigue behavior.

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TABLES

Table 1. Description of materials, commercial name, manufacturer, composition and batch number.

Material	Commercial name/manufacturer	Composition	Batch number
5% hydrofluoric acid	IPS Ceramic Etching Gel, Ivoclar Vivadent, Schaan, Liechtenstein	< 5% hydrofluoric acid	W14921
10% hydrofluoric acid	Condac Porcelana, FGM, Joinville, Brazil	< 10% hydrofluoric acid	211019
Epoxy resin	Protec, São Paulo, Brazil	Continuous filament woven fiberglass bonded with epoxy resin	-
Lithium disilicate ceramic	IPS e.max CAD, HT A2, C14, Ivoclar Vivadent	SiO_2, Li_2O, K_2O, P_2O_5, ZrO_2, ZnO, other and colouring oxides	W37404
Luting resin composite	Multilink N, Ivoclar Vivadent	Silicate glass, ytterbium trifluoride, highly dispersed silica, catalysts and stabilizer, pigments	W44613
Primer	Multilink N Primer A and B, Ivoclar Vivadent	Primer A: water, initiators; Primer B: phosphoric acid acrylate, hydroxyethyl methacrylate, methacrylate mod. polyacrylic acid, stabilizer	Primer A: Y25800 Primer B: Z003PX
Silane-based coupling agent	Monobond N, Ivoclar Vivadent	Alcohol solution of silane methacrylate, phosphoric acid methacrylate and sulphide methacrylate	Y19262

*The chemical composition is described according to the manufacturers' information.

Table 2. Experimental design.

Group	Surface Treatment	Storage regimen
CA	Silane-based coupling agent application for 15 seconds actively and 45 seconds passively	Baseline*
CA_AG		Aging**
HF	5% HF acid etching for 20 seconds	Baseline
HF_AG		Aging
HF+CA	5% HF acid etching for 20 seconds + silane-based coupling agent application for 15 seconds actively and 45 seconds passively	Baseline
HF+CA_AG		Aging

* 1 up to 5 days of water distilled storage (37ºC);

** 180 days of storage in distilled water + 25,000 thermal cycles, considered as long-term aging.

Table 3. Mean fatigue failure load (in Newton), number of cycles for failure with respective confidence interval, the percentage of decrease comparing the baseline condition to aging in both outcomes and de percentage of failure mode in each treatment.

Group	Fatigue failure load		% of mean FFL decrease (baseline/aging)	Cycles for failure		% of mean CFF decrease (baseline/aging)	% Failure Mode
	Baseline	Aging		Baseline	Aging		
CA	893 (774 – 1012) [b]	639 (534 – 744) [c]	28%	143,667 (119,863 – 167,470) [b]	84,179 (61,530 – 106,828) [c]	41%	Baseline: 100% radial crack Aging: 27% debonding
HF	1247 (1175 – 1318) [a]	817 (754 – 879) [b]	34%	214,333 (200,088 – 228,579) [a]	128,333 (115,839 – 140,827) [b]	40%	100% radial crack
HF+CA	1290 (1205 – 1375) [a]	900 (859 – 941) [b]	30%	222,333 (205,827 – 238,839) [a]	145,000 (136,885 – 153,115) [b]	35%	100% radial crack

* Same lowercase letters on each column/row indicate statistical similarity depicted by Kaplan Meier and Mantel-Cox post-hoc test.

Table 4. Weibull modulus for fatigue failure load and cycles for failure.

Group	Fatigue failure load		Cycles for failure	
	Baseline	Aging	Baseline	Aging
CA	4.50 (3.01- 6.72) [B]	4.15 (2.61 – 6.59) [B]	3.59 (2.39 – 5.39) [B]	2.12 (1.33 – 3.35) [B]
HF	12.35 (8.05 – 18.95) [A]	8.04 (5.39 – 11.99) [AB]	10.58 (6.89 – 16.25) [A]	6.33 (4.23 – 9.46) [AB]
HF+CA	9.23 (6.20 – 13.75) [AB]	15.37 (10.18 – 23.2) [A]	8.19 (5.50 – 12.21) [AB]	12.36 (8.18 – 18.69) [A]

* Same uppercase letters on each column/row indicate statistical similarity based on maximum-likelihood estimations for Weibull analysis.

FIGURES

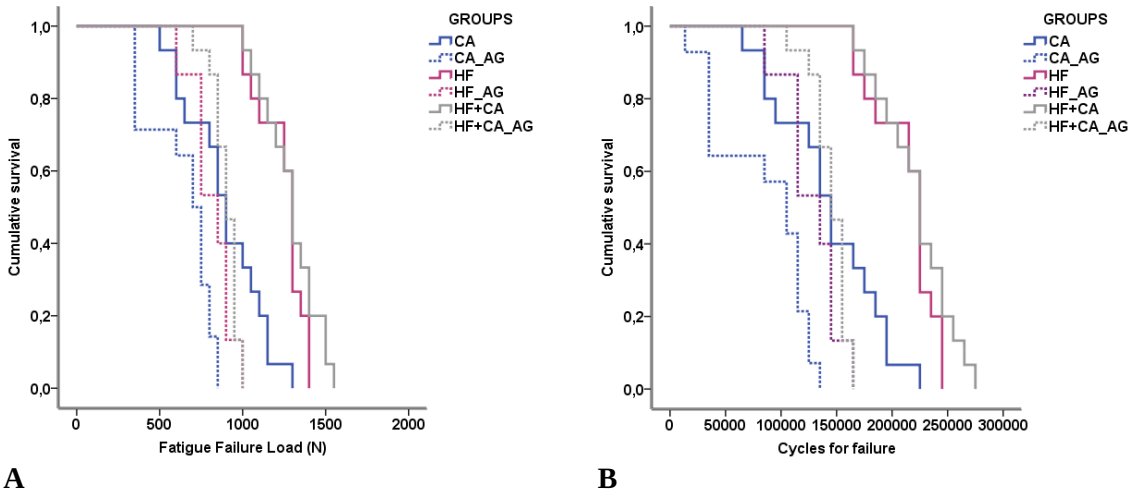


Figure 1. Survival graphs obtained by the Kaplan-Meier and Mantel-Cox (log-rank) test: 'A' for fatigue failure load in Newton and 'B' for the number of cycles for failure, clearly showing the impairment in the fatigue performance (load and cycles) of all groups after aging (dotted lines). Distinct colors demonstrate difference among different treatments.

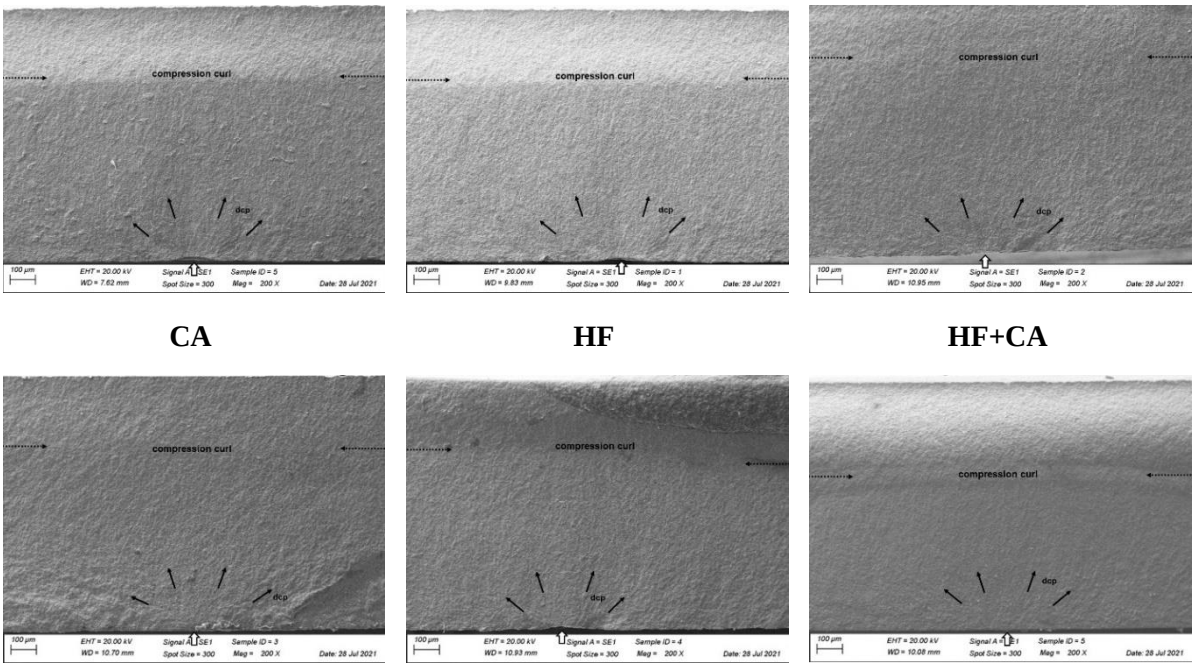
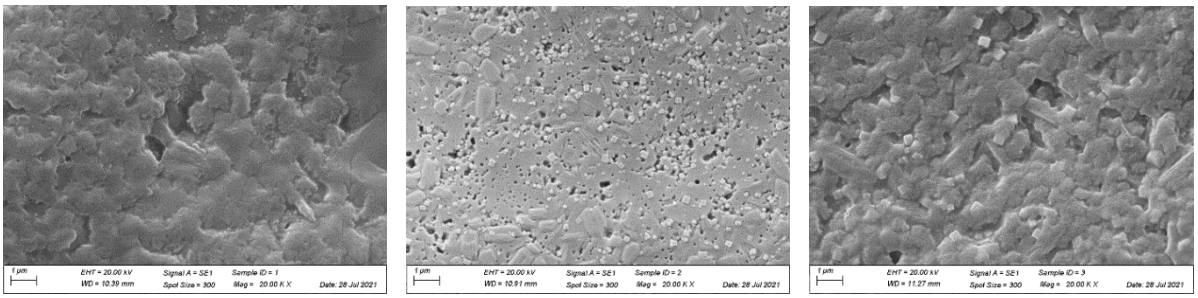


Figure 2. Fractographic images (200×) obtained by SEM illustrating that failures originated at cementation surface from defects present at the ceramic surface, pointed by white arrows, and then propagated to the top surface (direction of crack propagation - dcp), where it can be noticed the compression curl.



CA **HF** **HF+CA**

Figure 3. Topographic images (20,000×) obtained by SEM show the CA group (non-etched ceramic surface) with a smoother and more regular surface; HF group with an evident glass matrix dissolution, exposure of crystallographic intergranular regions and creation of micro retentive grooves; HF+CA with a more homogeneous surface compared to HF alone, leading to the understanding that the silane-based coupling agent filled and smoothed surface defects.

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3 ARTIGO 2: Adhesion to lithium disilicate glass-ceramics after aging: Resin viscosity and ceramic surface treatment effects

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Adhesion to lithium disilicate glass-ceramics after aging: Resin viscosity and ceramic surface treatment effects

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Running title: Resin cement viscosity on the bond strength of ceramic

Adhesion to lithium disilicate glass-ceramics after aging: Resin viscosity and ceramic surface treatment effects

Abstract

Objective: To evaluate the influence of intaglio ceramic surface treatments, resin cement viscosities, and storage regimens on the microshear bond strength of lithium disilicate ceramic. In addition, to investigate the dynamic viscosity of the resin-based luting agents.

Materials and Methods: Ceramic slices were randomly allocated into eight groups (n= 19) considering three factors: ceramic surface treatment (hydrofluoric acid followed by silane, HF; or self-etching ceramic primer, E&P), resin cement viscosity (high, HIGH; or low, LOW) and storage regimen (baseline or aging). Surface treatments were performed, resin cement cylinders were built and microshear bond strength tests (μSBS, wire-loop method, speed: 1.0 mm/min) were run according to the storage factor. Failure mode, topographic and dynamic viscosity (37°C; shear rate of 1.0 – 100 s^{-1}) of resin cement components (base, high and low catalyst) were also performed.

Results: Resin cement viscosity and the association among ceramic surface treatment, resin cement viscosity, and storage regimen were statistically significant factors ($p < 0.05$). Worse behavior was identified for the E&P_HIGH group compared to the E&P_LOW and HF_LOW in the baseline condition. After aging, the HF_HIGH group (16.78 MPa) presented the worst result among the aged groups (21.44 – 25.25 MPa). Most of the failures were adhesive. Surface micrographs revealed a distinct pattern after etching, more aggressive by HF and milder by E&P. High viscosity catalyst is 5.3 and 8.5-fold more viscous than the base and low viscosity catalyst, respectively (high > base > low).

Conclusion: Differences in filler content can impact the resin viscosity of the material (more fillers increase the viscosity), which in turn can influence the bond strength of a lithium disilicate ceramic, depending on the surface treatment and storage regimen.

Keywords: Ceramics. Micro-shear. Rheology. Resin composite cements.

Highlights

- High and low resin cements components show different dynamic viscosities;
- Viscosities of resin cement affecting the resin adhesion to glass-ceramic;
- Aging only negatively affects HF etched lithium disilicate bonded with high viscosity.

1. Introduction

Lithium disilicate glass-ceramic is well-known for being versatile and recommended for multiple clinical applications in both occlusal stress and esthetic demand areas (Aziz et al., 2020; Bacchi and Cesar, 2022). The production of precise ceramic restorations has been achieved by using technologies such as computer-aided design and manufacturing (CAD-CAM) and improvements in these directions are likely to increase the predictability of dental treatments based on ceramic restorations (Bacchi and Cesar, 2022). On the other hand, milling, fitting adjustments, and etching are procedures that can introduce defects on the intaglio surface of lithium disilicate restorations and may affect the mechanical and surface properties (May et al., 2021). Considering that ceramics are brittle, the intaglio-damaged surface is a critical zone (Alves et al., 2022), but this can be minimized by adhesive bonding (da Rosa et al., 2022).

Among the key principles of a long-lasting and stable adhesion is to generate a rough surface for interfacial interlocking, good surface wetting, and appropriate adaptation for intimate interaction (Marshall et al., 2010). Therefore, the intaglio surface of the ceramic needs to be subjected to a conditioning protocol after restoration processing. According to a recent systematic review and meta-analysis (Lima et al., 2022), an appropriate surface treatment protocol for glass-ceramics is based on two main strategies: mechanical, to develop micromechanical interlocking; and chemical, to link the glass-ceramic material to the resin cement. For this purpose, a self-etching ceramic primer (etching/priming) appears to be an efficient substitute for traditional hydrofluoric acid followed by silane application in bond strength outcomes, simplifying the clinical bonding procedure (Tribst et al., 2018; Dapieve et al., 2021; Lima et al., 2022).

The other key adhesion principle mentioned by Marshall et al. (2010) is the adequate viscosity of the adhesive system. In a resin bonding context, the luting agent should penetrate the surface defects, sealing the microcracks, and as a result reinforce the restorative set (da Rosa et al., 2022). An ideal resin-based viscosity allows minimal thickness, ease of placement on the restored site, and proper interpenetration of the milled/treated ceramic. The viscosity can be modified by preheating the luting agent (Marcondes et al., 2020; Barbon et al., 2022), changing the proportion of the resin matrix (Lee et al., 2006; Barbon et al., 2019; Dapieve et al., 2022), using different compositions (Lee et al., 2006; Beun et al., 2009; Coelho et al., 2019) or by distinct inorganic filler sizes and morphologies (Lee et al., 2006). Finally, regardless of how the viscosity is changed, it might affect the wettability of ceramic surfaces (Barbon et al., 2019), and thus influence the adhesion.

The ultimate goal of good adhesion aims to produce an interface that is strong and durable. Understanding the interfaces against challenges and their defects over time as a function of water effects and thermal cycling is important to improve the predictability of the restorative bonds (Marshall et al., 2010). Therefore, laboratory procedures for long-term aging of the interface are strongly encouraged (Armstrong et al., 2017; Van Meerbeek and Frankenberger, 2020). Studies that evaluated the bond strength between glass-ceramics and resin cements at different viscosities did not subject the specimens

to an aging regime, and knowing the importance, they highlighted this drawback in their limitations (Barbon et al., 2019; Dapieve et al., 2022).

Based on the importance to evaluate resin-based viscosities bonded to glass-ceramic treated by different strategies and submitted to a durability challenge, this study aimed to evaluate the influence of the intaglio ceramic surface treatments (hydrofluoric acid followed by silane application or self-etching ceramic primer), the resin cement viscosity (high or low) and the storage regimen (baseline or aging) on the microshear bond strength of lithium disilicate ceramic. Furthermore, the dynamic viscosity of the resin cements components was also investigated. The hypotheses were: (1) the ceramic surface treatments and the resin cement in different viscosities will induce similar bond strength; (2) aging will have a detrimental effect on the bond strength; and (3) the more filled particle resin cement will present higher apparent viscosity.

2. Material and methods

A general description of the materials used in the present study is listed in Table 1.

2.1. Study design

A sample size calculation considering the resin cement cylinder as a sample unit was performed (OpenEpi, Dean et al., 2006) considering the results of a microshear bond strength pilot study. A confidence interval of 95%, a statistical power of 80%, and a difference of 3.5 between high viscosity resin cement (mean= 17.62; standard deviation, SD= 3.85) and low viscosity resin cement (mean= 14.12; SD= 3.81) at baseline condition were considered, resulting in 19 resin cement cylinders per group.

Accordingly, the experimental design of this study (Fig. 1) consisted of eight groups (n= 19), following the factors:

i. “Ceramic surface treatment”: 5% hydrofluoric acid followed by silane-based coupling agent or self-etching ceramic primer;
ii. “Resin cement viscosity”: high or low viscosity;
iii. “Storage regimen”: baseline or aging;

2.2 Specimen preparation

Lithium disilicate glass-ceramic pre-crystallized CAD-CAM blocks (IPS e.max CAD LT A2/C16, Ivoclar, Schaan, Liechtenstein) were sectioned into rectangular slices (16 mm x 9 mm x 2 mm) using a diamond disc at low speed under water cooling in a cutting machine (Isomet 1000, Buehler, Lake Bluff, United States). The ceramic bonding surfaces were polished (EcoMet/AutoMet 250, Buehler) in a sequence of #400-, #600-, and #1200-grit silicon carbide papers (SiC) to produce a standardized polished surface and eliminate cut scratches. As in-lab simulation methods of CAD-CAM (CEREC system) milled surfaces for specimen fabrication are capable of inducing statistically similar bond strength to the milled samples, a laboratory simulation with SiC papers was performed (Pilecco et

al., 2022). For this purpose, the ceramic bonding surfaces were submitted to a CAD-CAM in-lab simulation using a standardized size (100 mm x 50 mm) of #60-grit SiC humidified paper for each ceramic. The grinding was carried out with lightweight hand pressure for 15 s on each axis of the specimen (x and y axes by a single-trained operator (KSD) (Dapieve et al., 2021; Pilecco et al., 2022).

Then, the ceramic slices were crystallized according to the manufacturer's instructions (840°C, 7 min vacuum; Vacumat 6000 MP, VITA Zahnfabrik, Bad Säckingen, Germany), and the surface roughness of the ceramic samples was measured through the contact surface roughness (Mitutoyo SJ-410; Kanagawa, Japan) according to ISO 4287. Six measurements were performed on each specimen (x and y axes) considering the average roughness (Ra) and mean distance between the five highest peaks and valleys (Rz). The roughness values achieved by the CAD-CAM in-lab simulation (mean ± SD: Ra= 1.58 ± 0.29; Rz= 10.00 ± 0.98) were close to those generated by CAD-CAM machining (mean ± SD: Ra= 1.90 ± 0.08; Rz= 11.46 ± 0.41, Pilecco et al., 2022).

Next, the ceramic slices were embedded in polyvinyl chloride cylinders (PVC, Krona, Joinville, Brazil) with self-curing acrylic resin (JET Clássico, Campo Lindo Paulista, Brazil), leaving the simulated surface free for the ceramic surface treatments. The specimens were cleaned (isopropyl alcohol, 5 min) in an ultrasonic bath (1440 D, Odontobras, Ribeirão Preto, Brazil), and randomly allocated (www.random.org) into the 8 testing groups (Fig. 1).

2.3 Ceramic surface treatments

The bonding surfaces were treated according to the "ceramic surface treatment" factor, following the manufacturer's instructions and a previous study (Dapieve et al., 2021):

i. **5% hydrofluoric acid followed by a silane-based coupling agent (HF):** 5% HF (IPS Ceramic Etching Gel, Ivoclar) was applied with a micro applicator for 20 s, rinsed with air-water spray for 30 s, air-dried for 30 s and subsequently cleaned in an ultrasonic bath (distilled water; 5 min). A silane-based coupling agent (Monobond N, Ivoclar) was applied with a micro applicator for 60 s and gently air-dried.
ii. **Self-etching ceramic primer (E&P):** the E&P system (Monobond Etch & Prime, Ivoclar) was actively applied with a micro applicator for 20 s and allowed to react for a further 40 s. Then, the surfaces were rinsed with air-water spray for 30 s, air-dried for 30 s, and cleaned (ultrasonic bath, distilled water, up to 5 min).

2.4. Bonding procedures

After the surface treatments, starch matrices (1.13 mm internal diameter; 1.0 mm high; Isabela, M. Dias Branco S.A., São Caetano do Sul, Brazil) (Tedesco et al., 2013) were positioned over each treated surface and fixed with sticky wax (Lysanda, São Paulo, Brazil), with a total of nineteen (n= 19) matrices for each group.

A dual-curing resin cement (Variolink N, Ivoclar) in a high or in a low viscosity catalyst was proportioned in a 1:1 ratio with the base (high viscosity + base; low viscosity + base) and manipulated. Then, the resin cement was carefully inserted with a resin spatula (Millennium Titanium Resin Nº9, Golgran, São Caetano do Sul, Brazil) into each starch matrix according to the "resin cement viscosity" factor. The resin cement was light-cured (1200 mW/cm^2, Radii-Cal, SDI, Bayswater, Victoria, Australia) for 40 s and the samples were stored (24 h, distilled water, 37ºC). Then, the starch matrices were carefully removed with a dental explorer (#5, Hu-Friedy Manufacturing, Chicago, United States) to obtain the resin cement cylinders (sample unit). The specimens were individually assessed using an optical microscope (Stereo Discovery V20, Carl Zeiss, Göttingen, Germany) at 40× magnification to identify any failures (i.e. bubbles, porosities) at the adhesive interface. Failed samples were excluded and replaced in order to maintain the sample size.

2.5 Storage conditions

The specimens of each condition were randomly assigned into two following storage conditions before testing:

i. **Baseline**: storage in distilled water at 37°C for 24 h;
ii. **Aging**: storage in distilled water at 37°C for 120 days (medium-term, Armstrong et al., 2017) and 25,000 thermal cycles (Armstrong et al., 2017; Van Meerbeek and Frankenberger 2020) of 30 s baths at 5 and 55ºC with a transfer time of 5 s (Nova Ética, Vargem Grande do Sul, Brazil).

2.6 Microshear bond strength test (μSBS)

Each specimen was coupled to a dedicated shear device in a universal testing machine (EMIC DL-2000, EMIC, São José dos Pinhais, Brazil). The microshear test was performed using the wire-loop method (stainless steel wire, Ø= 0.20 mm, Tedesco et al., 2013; Prado et al., 2018; Prochnow et al., 2018). For testing purposes, the stainless-steel wire loop was positioned as close as possible to the adhesive interface, and a constant load at a crosshead speed of 1.0 mm/min was applied until failure. The microshear bond strength was calculated according to the formula: S= L/A; where "S" is the strength (MPa), "L" is the load recorded for the failure of the specimen (N), and "A" is the interface area of the resin cement cylinder (standardized size of 1.00 mm^2).

2.7 Failure mode and topographic analysis

The fracture pattern of all samples was determined under a stereomicroscope (Stereo Discovery V20, Carl Zeiss, Gottingen, Germany), and classified into two types, according to previous studies (Prochnow et al., 2018; Dapieve et al., 2021):

i. **Adhesive failure**: failure at the interfacial region between the resin cement and ceramic (adhesive areas in more than 50% of the interface area);

ii. **Cohesive failure**: cohesive failure of the resin cement (cohesive areas in more than 50% of the interface area).

Then, representative fractured specimens (adhesive and cohesive; n= 1) were ultrasonically cleaned (distilled water, 5 min), gold-sputtered and evaluated under Scanning Electron Microscopy (SEM, Vega3, Tescan, Brno, Czech Republic) to observe the failure pattern at 150× magnification.

Additional specimens of each "ceramic surface treatment" factor (HF and E&P; n= 1) were also ultrasonically cleaned, gold-sputtered, and analyzed under SEM at 20,000× magnification to observe the surface alterations of the treatments.

2.8 Dynamic viscosity analysis

Dynamic viscosity analysis was performed based on a previous study (Marcondes et al., 2020) to characterize the viscosity of each resin cement component (base, high, and low viscosity catalysts). The measurements were carried out using a stress-controlled MCR 102 rheometer (Anton Paar, Modular Compact Rheometer, Ostfildern, Germany), equipped with a cone-plate geometry (cone diameter= 25 mm, angle= 1° with 0.058 mm gap) under atmospheric pressure. The shear rate test was done in the range of 1.0 - 100 s^{-1} at a controlled temperature (37°C) by using a Peltier plate. This temperature was used to simulate the clinical condition after seating the restoration. The resin cement components were placed on the lower plate of the rheometer (< 0.2 ml), and after positioning the resin between the plates, the excess sample was removed before viscosity measurements. The analysis was performed in triplicate (n= 3) and 30 points were determined in each shear rate test. The RHEOPLUS/32 version 3.62 software program (Anton Paar) was used to obtain the viscosity, following the equation:

$$\text{viscosity} = \frac{shear\ stress}{shear\ rate} \rightarrow \ \eta = \frac{\tau}{\dot{\gamma}}\ [\text{Pa.s}] = [\frac{Pa}{s-1}]$$

2.9 Statistical analysis

A statistical software program (IBM SPSS Software, IBM, Armonk, United States) was used to perform the analysis with a significance level of 0.05, discarding the cohesive failures, since they do not represent the real bond strength value (Wang and Duong, 2016). Bond strength data were diagnosed as non-normal by the Shapiro-Wilk test (SW, p= 0.042) and homoscedastic by Levene's test (p= 0.798). A factorial analysis of variance (ANOVA, 3x2) was performed to compare the "ceramic surface treatment*resin cement viscosity*storage regimen". Due to non-normal data, bootstrapping procedures (1000 re-samplings; 95% CI, bias-corrected and accelerated, BCa) were implemented to obtain greater reliability of the results, to correct deviations from the normality of the sample distribution, and to present a 95% CI for differences among means (Haukoos and Lewis, 2005).

Failure mode and topographic SEM were qualitatively evaluated. The viscosity data of three resin cements components at a constant shear rate of 2 s^{-1} was diagnosed as non-normal by SW (p=

0.001) and non-homoscedastic by the Levene's test (p= 0.044). One-way ANOVA and Bonferroni's post-hoc test was performed using bootstrapping procedures (1000 re-samplings; 95% CI, BCa).

3. Results

μSBS data are displayed in Tables 2 (F and p values) and 3 (mean and standard deviation, samples tested, and type of failure).

Three-way ANOVA showed a statistically significant effect for resin cement viscosity (F= 10.876, p= 0.001) and for the ceramic surface treatment*resin cement viscosity*storage regimen (F= 9.708; p= 0.002). Multiple comparisons revealed worse behavior for the E&P_HIGH group compared to the E&P_LOW and HF_LOW in the baseline condition. After aging, only the HF_HIGH group had a significant reduction in bond strength performance, presenting the worst result among the aged groups.

Only one pre-test failure occurred (E&P_LOW aging, after the thermal cycles) considering all specimens, and most of the failures were adhesive (Table 3).

SEM analysis showed the difference between failure modes, showing a circle boundary in the adhesive failure (starch matrices shape) and a gathering of the resin cement in the cohesive failure (Fig. 2). Surface micrographs revealed a distinct pattern after etching, more aggressive by HF and milder by E&P (Fig. 3).

The profile of shear stress for the base, high viscosity, and low viscosity catalyst at 37°C as a function of shear rate is shown in Fig. 4. The flow curves of the three components of resin cements show a decreasing curve slope, indicating a shear-thinning (pseudoplastic) behavior (i.e. the apparent viscosity decrease with the shear rate increases). The statistics indicate that apparent viscosity at a constant shear rate of 2 s^{-1} is a significant factor (p= 0.000; F= 957.069). High viscosity catalyst is 5.3 and 8.5-fold higher viscous than the base and low viscosity catalyst, respectively (high > base > low, Table 4).

4. Discussion

The differences in viscosity detected between the evaluated resin cements components (higher viscosity for the high catalyst and lower viscosity for the low catalyst) had an impact on the microshear bond strength. Lithium disilicate treated with hydrofluoric acid followed by silane presented the best performance with low viscosity, meanwhile, the viscosity of the resin cement used for the ceramic treated with a self-etching ceramic primer is indifferent. On the other side, ceramic surface treatments (HF and E&P) and storage conditions (baseline and aging) did not influence the adhesion outcome. Therefore, the first hypothesis that ceramic surface treatments and resin cement at different viscosities would induce similar bond strength; and the second hypothesis, that aging would have a detrimental effect on bond strength, were both rejected. The third hypothesis, that cement resin with more fillers would have a higher apparent viscosity, was accepted.

The main findings of this study are related to the resin cement viscosity regarding the verification of the viscosity differences (Table 4, Fig. 4) and its influence on the bond strength to lithium disilicate (Table 3). One of the limitations reported by a previous article, which also evaluated different viscosities of resin cement and bond strength to lithium disilicate (Dapieve et al., 2022[a]), was the definition of high and low viscosity based on the composition provided by the manufacturer (variations in the percentage of inorganic fillers, Table 1: high 77.2% and low 71.2% wt.). Thus, in the present article, we confirmed that high and low catalysts present different viscosities at body temperature (37ºC, high > low; Table 4). These viscosity changes are in accordance with the premise that the higher the percentage of inorganic fillers, the greater the viscosity (Lee al., 2006; Barbon et al., 2019). The mechanism of the increased viscosity by increasing the number of fillers can be explained by the inorganic content having a higher stiffness than the polymer matrix. In this sense, the increase in viscosity is a direct consequence, since more fillers mean a lower interparticle distance, affecting the flowability of the unpolymerized material (Barbon et al., 2019).

Another relevant aspect of the resin cement viscosity is that as the shear rate increases, the viscosity decreases, regardless of the percentage of inorganic fillers (Table 1, Fig. 4). This decreasing phenomenon is due to the array of particles becoming directional and the interaction between them decreases, which is characteristic named shear thinning (pseudoplastic behavior). Extrapolating this to clinical terms, when we manipulate the resin cement, it tends to have greater fluidity due to that behavior, which is a positive point, as it facilitates the handling characteristics (Lee et al., 2006).

Having defined what is meant by the resin cement viscosity, it is important to consider the interpenetration of those luting agents in the etched surface patterns (Fig. 3). Clearly, the ceramic surface with CAD-CAM in-lab simulation and etched by hydrofluoric acid is more porous and shows deeper voids compared to self-etching ceramic primer, in line with previous extensive research (Prado et al., 2018; Tribst et al., 2018; Dapieve et al., 2021; Dapieve et al., 2022[b]). This surface characteristic added to the resin cement viscosity characteristics and aging can explain some of the bond strength results (Table 2). The E&P and high viscosity presented the lowest bond strength values in the baseline condition (E&P_HIGH < E&P_LOW, Table 3), which can be explained by the difficulty of the resin cement to penetrate shallower defects, even though E&P_HIGH was also similar to HF_HIGH. Regarding the HF treatment, which presents wider and deeper defects, both resin cements were possibly able to fill them, resulting in a similar adhesive behavior (HF_HIGH = HF_LOW), in line with the findings of the study by Dapieve et al. (2022[a]).

After aging, the more pronounced HF etching defects seem to have been more critical, leading to intensified degradation of the adhesive interface, with the worst adhesive performance for the high viscosity cement (HF_HIGH < HF_LOW). Thus, the filler content of the resin cement (viscosity) appears as a significant factor (Table 2), as according to Barbon et al. (2019). Those authors also observed higher bond strength values for the resin-based luting agents with low and intermediate inorganic filler content (low and intermediate viscosity). This means that the low and intermediate

viscosity was fluid enough to fill the irregularities created on the ceramic by acid etching and to generate adequate micromechanical interlocking, but the more viscous material (high viscosity) was not sufficient (Barbon et al., 2019), especially considering a long-term aging evaluation as herein. Resin-based materials are not inert in the oral environment and may release components, initially due to incomplete polymerization, and later due to degradation (aging of composite materials may also lead to more porosities due to an interplay of mechanical swelling and water sorption and chemical degradation) (Van Landuyt et al., 2011). Even with an aggressive aging protocol, it is possible to note that the only one which did not show stability was the HF_HIGH when comparing baseline and aged groups. In this case, the pronounced defects from etching may have been critical, and adding the high resin cement viscosity which is increased by fillers can then suffer more swelling process and degradation (Niem et al., 2020).

Regarding the ceramic primer, it seems that the smoother topographical characteristic may allow similar micromechanical interlocking for both cements, which resulted in a similar adhesive behavior even after aging. In addition, the self-etching ceramic primer can generate stable adhesion to lithium disilicate ceramic (Prado et al., 2018; Dapieve et al., 2021; Tribst et al., 2021), explaining the stability of this surface treatment; however, there was no consensus on this premise, as some studies also reported a deleterious impact on bond strength after aging (Tribst et al., 2018; Dimitriadi et al., 2020).

A contradictory aspect that is important to mention concerns the results found here (influence of viscosity factor) compared to those of Dapieve et al. (2022[a]), which found a similarity between HF groups and high/low viscosity cements. This divergence of bond strength data can be explained by the distinctive ceramic surface characteristics (Pilecco et al., 2022). Despite both being treated with HF followed by silane, the surface processing in the present study was based on CAD-CAM in-lab simulation, with an initial average surface roughness of Ra= 1.58/Rz= 10.00. In Dapieve et al., the processing surface method was only polishing. The authors did not report the mean roughness after polishing, but in Pilecco et al. (2022) the roughness after the same polishing protocol was Ra= 0.03/Rz= 0.26. Considering that the bond strength found in a set is based on the micromechanical interlocking of the ceramic surface and resin cement interpenetration, this difference can be relevant. Despite polished specimens being useful for standardizing *in vitro* studies, they produce statistically underestimated bond strength results compared with milled and simulated specimens and should therefore be used with caution (Pilecco et al., 2022).

Despite the evidence obtained in this study, there are some drawbacks to be considered, such as the topography achieved by the CAD-CAM in-lab simulation. Even though the surface roughness values are similar, there may be undeniable differences in the shape, size, and quantity of the defects due to the inherent particularities of the processing method itself. The performed viscosity provides information for the different components of the resin cement and not for the final mixture, due to limitations inherent to the analysis. Also, the adhesive behavior considering dentinal and enamel substrates is strongly encouraged. Even so, our study is important to illustrate that differences in filler content can be decisive

for the resin viscosity of the material, which in turn can influence the bond strength of a lithium disilicate ceramic, depending on the surface treatment and aging.

5. Conclusions

- Differences in resin cement viscosities can affect microshear bond strength to lithium disilicate ceramics, depending on the ceramic surface treatment (HF/E&P) and storage regimen (baseline/aging);
- Aging only negatively damages the HF etched lithium disilicate bonded with high viscosity resin cement

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Tables

Table 1. Information about the materials used in the study: commercial name, manufacturer, composition according to the manufacturer's information, and batch number.

Material	Trade market and manufacturer	Main composition (% in weight)	Filler % wt.	Filler % vol.	Batch number
5% HF	IPS Ceramic Etching Gel, Ivoclar, Schaan, Liechtenstein	< 5% Hydrofluoric acid	-		W14921
Dual-curing resin cement	Variolink N High Viscosity (Catalyst), Ivoclar	Barium glass filler and mixed oxide (52.2%**), dimethacrylates (22.0%), ytterbiumtrifluoride (25%), initiators and stabilizers (0.8%) and pigments (< 0.1%)	77.2 %	52.0 %	Y15347
	Variolink N Low Viscosity (Catalyst), Ivoclar	Barium glass filler and mixed oxide (46.2%), dimethacrylates (27.9%), ytterbiumtrifluoride (25%), initiators and stabilizers (0.9%) and pigments (< 0.1%)	71.2 %	43.6%	Y06775
	Variolink N Base, Ivoclar	Barium glass filler and mixed oxide (48.4%), dimethacrylates (26.3%), ytterbiumtrifluoride (25%), initiators and stabilizers (0.3%) and pigments (< 0.1%)	73.4 %	46.7 %	Y06775
Lithium disilicate ceramic	IPS e.max CAD, Ivoclar	SiO_2, Li_2O, K_2O, P_2O_5, ZrO_2, ZnO, Al_2O_3, MgO, oxides	-		W37404
Self-etching ceramic primer	Monobond Etch & Prime, Ivoclar	Ammonium polyfluoride, silane system based on trimethoxypropyl methacrylate, alcohols, water, and colorant			Y27773
Silane-based coupling agent	Monobond N, Ivoclar	Alcohol solution of silane methacrylate, phosphoric acid methacrylate, and sulfide methacrylate			Z00DTK

Table 2. Three-way ANOVA for the factors and interactions considering F and p values. Statistically significant interactions are highlighted in bold.

Factors/interactions	F	P
ceramic surface treatment – surface	0.027	0.870
resin cement viscosity – viscosity	**10.876**	**0.001**
storage regime - storage	0.112	0.739
surface*viscosity	0.351	0.555
surface*storage	0.756	0.386
viscosity*storage	2.583	0.110
surface*viscosity*storage	**9.708**	**0.002**

Table 3. μSBS (mean and standard deviation) in MPa, total samples of pre-tested failures after aging regimen (PTF – pre-test failures), total samples tested, and types of failure.

Groups	μSBS		Samples tested before/after aging/PTF	Type of failure			
				Adhesive		Cohesive	
	Baseline	Aging		Baseline	Aging	Baseline	Aging
HF_LOW	21.13 (5.81) $_{ab}$	25.25 (7.05) ^{a}	19/19/0	100%	100%	0%	0%
HF_HIGH	21.95 (4.37) ^{a}	16.78 (6.17) ^{c}	19/19/0	100%	100%	0%	0%
E&P_LOW	22.92 (7.55) ^{a}	22.62 (6.20) $_{ab}$	19/18/1	95%	100%	5%	0%
E&P_HIGH	18.77 (4.65) ^{b}	21.44 (5.62) $_{ab}$	19/19/0	100%	95%	0%	5%

*Different lowercase letters indicate statistical differences reported by three-way ANOVA.

Table 4. Resin cements viscosities (mean and standard deviation) at a constant shear of 2 s^{-1}.

Resin cement component	Viscosity (Pa.s)
Base	45.73 (0.81) ^{b}
High viscosity catalyst	244.33 (11.59) ^{a}
Low viscosity catalyst	28.70 (0.40) ^{c}

*****Different lowercase letters indicate statistical differences reported by one-way ANOVA and Bonferroni´s test**.**

Figures.

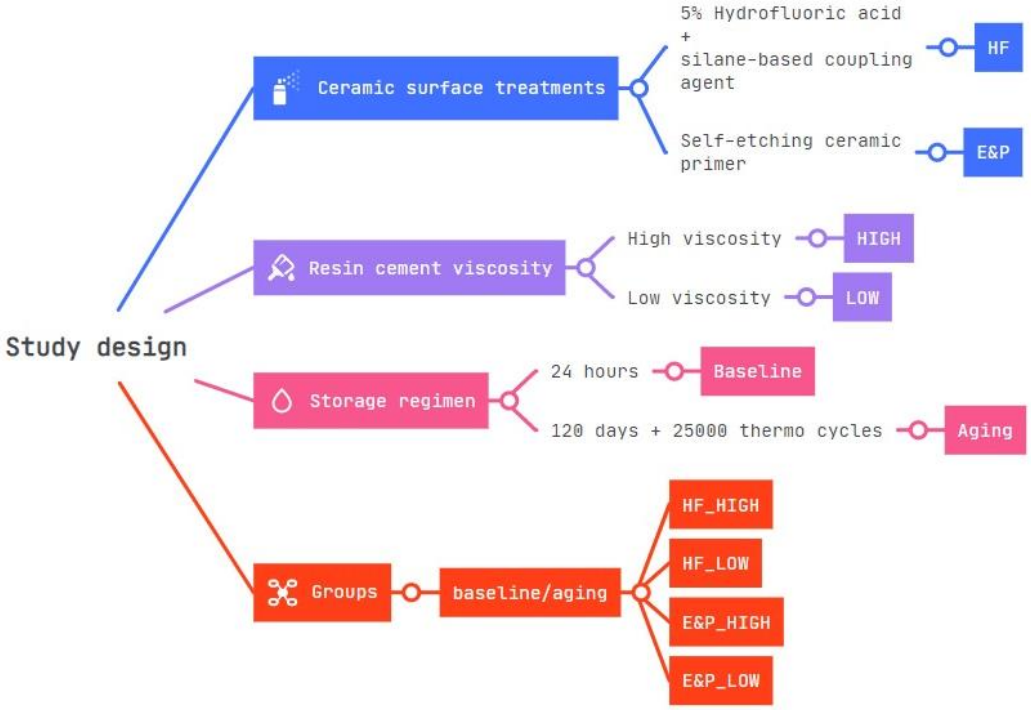
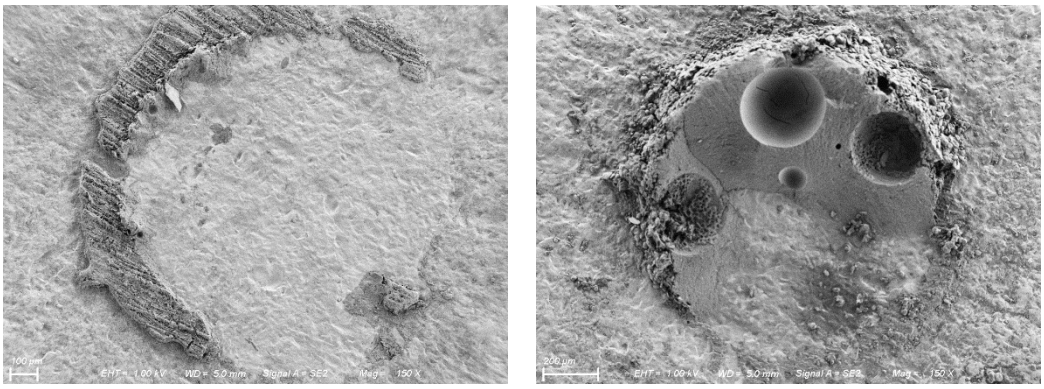
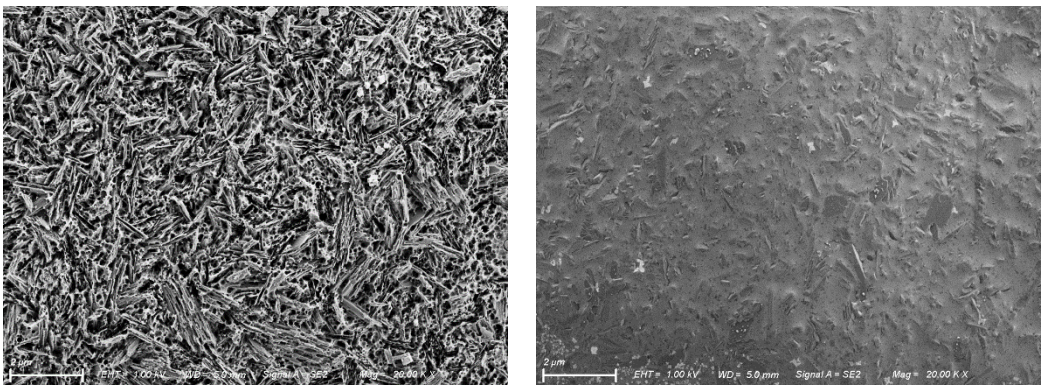


Figure 1. Experimental design (n= 19) for bond strength to lithium disilicate ceramic according to the 8 groups and 3 study factors: ceramic surface treatment, resin cement viscosity, and storage regimen.



A. **B.**

Figure 2. Representative SEM images of microshear tested specimens under 150× magnification in defined failure patterns: (**A**) adhesive and (**B**) cohesive failure at the resin cement.



A. **B.**

Figure 3. Representative micrographs under 20,000× magnification representing the topographic characteristics after CAD-CAM in-lab simulation and after each surface treatment. The HF etching (**A**) had an irregular topographic pattern with deeper defects compared to the E&P etching and priming (**B**), which had a shallower pattern.

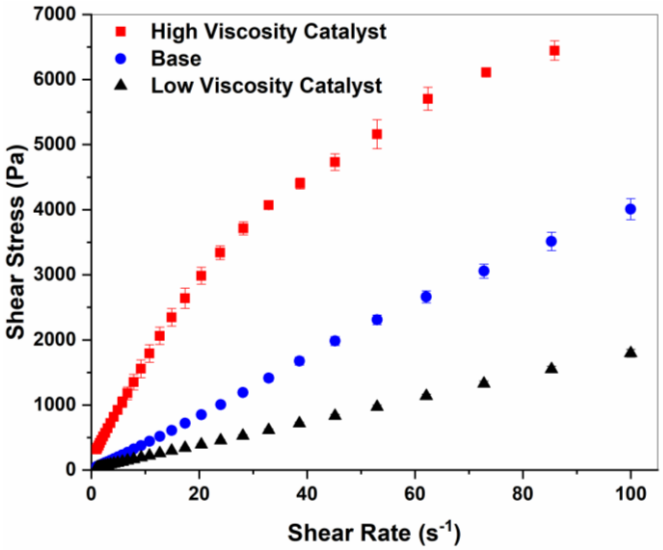


Figure 4. Shear stress versus shear rate for the base, high, and low viscosity catalyst at 37°C. The flow curves of the three resin cements components show that the apparent viscosity decrease as the shear rate increases. The apparent viscosity for high viscosity catalyst (red squares), base (blue dots) and low viscosity catalyst (black triangles) were statistically different ($p < 0.05$).

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4 ARTIGO 3: Cyclic fatigue vs static loading for shear bond strength test of lithium disilicate and dentin substrates: A comparison of resin cement viscosities

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Cyclic fatigue vs static loading for shear bond strength test of lithium disilicate and dentin substrates: A comparison of resin cement viscosities

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Running title: Static and fatigue shear bond strength

Abstract

Objective: To explore the effect of resin cement viscosities on the shear bond strength under static and fatigue load of lithium disilicate and dentin substrates.

Methods: Bonded tri-layer samples (lithium disilicate ceramic cylinder, resin cement, and substrate – ceramic or dentin) was performed considering 2 factors (n = 15): "resin cement viscosity" (high, HV; or low, LV) and "loading mode" (static, s-SBS; or fatigue shear bond strength, f-SBS). The specimens were subjected to s-SBS (1 mm/min, 1 kN load cell) and f-SBS (cyclic fatigue, initial load: 10 N; step-size: 5 N; 10,000 cycles/step; underwater). Failure mode, topography, and finite element analysis (FEA) were performed.

Results: The resin cement viscosity did not influence the s-SBS and f-SBS of lithium disilicate substrate; however, it affected the bond strength to dentin, with HV presenting the worst fatigue behavior (f-SBS = 6.89 MPa). Cyclic loading in shear testing induced a notorious detrimental effect with a relevant decrease (16–56 %) in bond strength and survival rates, except for dentin substrate and LV. Most failures were adhesive. A distinct pattern comparing the disilicate and dentin was identified and FEA demonstrated that there was a stress concentration on the top of the cement layer.

Significance: Cyclic fatigue loading in shear testing has detrimental effects on the adhesive behavior and survival probabilities of bonded lithium disilicate sets, regardless of resin cement viscosity. In contrast, resin cement viscosity affects the bond strength and the survival rates of dentin substrate submitted to cyclic loading mode, in which a low viscosity results in better performance.

Keywords: Adhesion. Cementation. Ceramics. Dental bonding. Dental cements. Glass-ceramic. Shear strength. Viscosity.

Highlights

- Shear bond strength under fatigue loading and survival rates of luted ceramic sets.
- The influence of cyclic loads on the impairment of the resin adhesive interface.
- Resin cement viscosity did not affect the shear under static and fatigue loading of ceramic sets.
- Resin cement viscosity affected the bond strength of dentin and lithium disilicate.

1. Introduction

Resin bonding plays an important role in addressing the issue of strengthening dental restorations [1,2]. The bonding strategy of glass-ceramics involves multiple technique-sensitive steps, mainly ceramic surface treatments, resin-based luting, and substrate conditioning [3,4]. Its goals are to promote great stress distribution over the restoration [5], to increase the fracture resistance [2], and to achieve a long-lasting bond between ceramic, resin cement, and substrate [3,6].

Before bonding glass-ceramics, treating the adhesive surface for creating topographical alterations and providing mechanical interlocking with the resin-based cement is required for bond and mechanical strength [7–10]. When acid etching is applied to the ceramic, sharp line angles are created by the glass and crystal phases are dissolved at differing rates. These surface irregularities may act as stress concentration areas during loading, introducing critical defects in the material [7]. In view of this, resin cement aims to infiltrate and seal microcracks at the ceramic surface, reducing the flaw population and increasing the energy required for catastrophic failure in a mechanism that strengthens the ceramic structure [5,7–9,11]. This means that adhesive bonding was able to overcome the strength degradation caused by acid etching and further improve the mechanical strength of the bonded structure [7,8].

Although resin cement infiltration of surface defects may act to reduce crack propagation, the luting agent is not always able to penetrate all defects, and the presence of unfilled porosities or flaws could concentrate stresses and reduce the overall strength [7,12]. Deficient infiltration may occur due to differences in resin cement viscosity, which can affect the ability of resin cement to flow over the surface [8,9,13]. A previous study of lithium disilicate glass-ceramic crowns demonstrated that luting strategies combined with hydrofluoric acid etching performed better when bonded with a high viscosity resin cement (77.2 % by weight of the catalyst paste) [13]. As already known, etching with 5 % hydrofluoric acid for 20 s resulted in an irregular topographic pattern with more profound and pronounced defects promoted by the dissolution of the glass matrix. The authors also observed that the high viscosity resin cement better filled these defects created by the acid. Also in this sense, strengthening may be dependent on the behavior of the resin inter-penetrating the ceramic surface, as reported by Addison et al. [1]. The strength and performance of resin-bonded ceramic restorations can be enhanced by the use of higher elastic modulus cements [1]. In fact, the set performance depended on the existing topography, the type of interfacial defect after surface treatment, and how the luting agent filled these defects [13].

From a clinical point of view, the resin cement must also have ideal viscosity to maintain a minimum layer thickness, allowing complete seating of the restoration without excessive pressure and avoiding stress concentration in the ceramic structure [5,14]. In addition, it has already been reported that the viscosity of the luting material determines the amount of displacement and occlusal surface height increase, as more fluid luting materials can avoid the risk of displacement and reduce the increase in occlusal surface height of the bonded restoration and thus minimize the need for occlusal adjustments and subsequent polishing [15].

The resin bond is also challenging considering a complex environment in the oral cavity where the adhesive interfaces are exposed to occlusal forces that consist of shear, tensile, compressive, and flexural stresses [3]. In addition, the restorations are continuously loaded, meaning that they are submitted to cyclic fatigue, a clinically relevant approach that produces the best insight into the material's response for its complete service life [16,17]. Some accelerated cyclic fatigue approaches use an increased step-by-step after a certain period (number of cycles) until failure or end of the test. This method considers the cumulative effect of the applied stresses, enabling the assessment of survival rates over time [16–18]. It is not possible to mimic the overall complexities of forces through an in vitro study; however, the results of these studies are important to help predict long-term performance [4,19,20].

As has been pointed out by researchers [21–23], static bond strength approaches do not reflect the dynamic loading of adhesive interfaces intraorally (i.e., the mode of these tests does not correspond to the type of forces encountered in the mouth). Restorative materials are submitted to cyclic forces that occur during oral function which result in failure by fatigue, constituting a different mechanism than that caused by static tests [21]. In fact, fatigue should be a central concern in the development of new dental materials and in assessing the success of restorative practices [22]. Although extensive research about resin bonds has been carried out, few studies have evaluated the adhesive interface challenged by a shear regime in cyclic fatigue loading (shear bond strength under mechanical fatigue) [21,22].

Lastly, "ceramic – resin cement – ceramic" restorative assemblies (respective ceramic surface treatments and luting procedures) are reported as a methodological alternative for bond strength tests and allow the factors to be isolated and evaluated separately [24,25]. However, taking into account that the adhesive success of the restoration is not only restricted to the bond strength of the resin cement to the ceramic, but also to the substrate to which the set adheres, approaches using dentin have great clinical relevance and should be evaluated [4,24,26,27]. Indeed, different substrate characteristics and microstructures can influence bond strength and reliability [5], and dentin is a critical substrate for adhesion; furthermore, it is still a challenge to understand how resin cement with different viscosities will act on hard tissues with low mineral content and a high proportion of organic compounds [27].

Based on these assumptions, this study should provide answers to gaps in the literature by exploring the influence of resin cement viscosities on shear bond strength under cyclic fatigue and survival rates of lithium disilicate glass-ceramics and dentin substrates when subjected to a cyclic loading regimen. Therefore, the objective of this study was to evaluate the influence of the resin cement viscosity (high or low) on the shear bond strength with different loading modes (static loading, s-SBS; or cyclic fatigue loading, f-SBS) on lithium disilicate ceramic, considering different substrates (lithium disilicate or dentin). The hypotheses were: (1) different viscosities of resin cement will induce similar shear bond strength; and (2) the fatigue loading mode will have a detrimental effect on the shear bond strength results on both substrates.

2. Material and methods

The information of the materials used in this study is listed in Table 1.

2.1. Study design

A sample size calculation was performed (OpenEpi) [28] considering the results of a static shear bond strength pilot study. A confidence interval of 95%, a statistical power of 80%, and a difference of 11.78 between high viscosity resin cement (mean= 30.75; standard deviation, SD= 14.87) and low viscosity resin cement (mean= 18.97; SD= 4.53) were considered. The minimum required sample was 14 specimens per group. Considering the possibility of a pre-test failure during the study progress, one specimen was added to each group, totaling 15 specimens per group (n= 15). The experimental design of this study is composed of four groups for each substrate (ceramic or dentin), following the factors (Table 2):

(i) "Resin cement viscosity": high or low viscosity;

(ii) "Loading mode": shear bond strength under static loading (s-SBS); or cyclic loading (fatigue approach, f-SBS).

A tri-layer bonded setup (ceramic cylinder - resin cement - substrate) was used, which is represented in a schematic drawing (Fig. 1A).

2.2 Specimen preparation

Lithium disilicate CAD/CAM blocks (IPS e.max CAD, Ivoclar, Schaan, Liechtenstein) were shaped into cylinders (Ø= 6 mm – substrate; Ø= 4 mm – restoration) using a diamond drill (Diamant Boart, Brussels, Belgium) coupled to a bench drill (SBE 1010 Plus, Metabo, Nürtingen, Germany), underwater cooling. Next, the cylinders were cut underwater cooling (Isomet 1000, Buehler, Lake Bluff, United States) in order to obtain 60 ceramic discs of substrates and 120 cylinders to be bonded onto the substrates (60 for ceramic substrate and 60 for dentin substrate). They were polished (#400-, #600-, and #1200-grit SiC papers, CarbiMet SiC Abrasive Paper, Buehler) in a polishing machine (Ecomet, Buehler) to standardize the surfaces and achieve a thickness of 1.5 mm for both substrate and cylinder. Then, the ceramic specimens were crystallized in a specific furnace (Programat P100, Ivoclar) according to the manufacturer's instructions (10 min at 840 °C, 7 min vacuum).

For the dentin substrate preparation, bovine tooth incisors [29] were cleaned and stored in 0.5 % chloramine at 4 °C. The root was cut (Ecomet, Buehler) at the cementoenamel junction and the crown was drilled (SBE 1010 Plus, Metabo) on the buccal surface into a cylinder format, resulting in 60 dentin substrate discs (Ø = 6 mm), one specimen from each bovine tooth. Then, the discs were polished (#400-, #600-, and #1200- grit SiC papers, CarbiMet SiC Abrasive Paper, Buehler) in a polishing machine (Ecomet, Buehler) to create a flat dentin surface and achieve a thickness of 1.5 mm. The substrates (ceramic and dentin) were subsequently coupled to a specific metallic shear device for the shear bond strength test (Fig. 1B).

2.3 Surface treatments and bonding procedure

The coupled dentin substrates and the ceramic specimens (substrates and cylinders) were cleaned in an ultrasonic bath (distilled water; 5 min) and carefully air-dried before the bonding procedure.

The bonding surface of the ceramic discs was etched with 5% hydrofluoric acid (HF) (IPS Ceramic Etching Gel, Ivoclar) for 20 s, rinsed with air-water spray for 30 s, air-dried for 30 s, and subsequently cleaned in an ultrasonic bath (distilled water; 5 min). Then, the silane-based coupling agent (Monobond N, Ivoclar) was applied for 60 s.

The bonding surface of the dentin substrate was treated with 35 % phosphoric acid (K-etchant syringe gel, Kuraray Noritake Dental Inc., Okayama, Japan) for 15 s, rinsed with air-water spray for 30 s, and carefully air-dried for 30 s. The adhesive of the luting composite Variolink system (Dualcuring total-etch adhesive ExciTE F DSC, Ivoclar) was applied for 10 s according to the manufacturer's recommendation.

Adhesive tapes (Scotch Magic Tape, 3M, Saint Paul, United States) were positioned precisely 1 mm from each other on the bonding surface of the substrates using a digital caliper (Absolute digimatic, Mitutoyo, Kawasaki, Japan), in order to restrict the bonding area as demonstrated in the schematic illustration (Fig. 1C). With the tapes in place and with the bonding surface untouched, the resin cement (Variolink N, Ivoclar) in high or low viscosity catalyst and base were mixed at a 1:1 ratio and applied on a ceramic restoration surface. The high and low viscosity of resin cement was followed by the manufacturer's scientific documentation, based on variations in the proportion (% weight, % volume) of the inorganic fillers (see Table 1). Each restoration was positioned on a specific substrate (dentin or lithium disilicate) according to the experimental design, and the assembly was seated under a static load of 100 g perpendicular to the adhesive interface. The resin cement excess was removed with a micro-applicator and the assemblies were light-cured (Elipar FreeLight 2, 3M) for two exposures of 20 s (Fig. 1D). Then, the coupled sets were matched with their corresponding analog metallic pair before the mechanical tests (Fig. 1E).

Next, additional lithium disilicate substrates for each resin cement viscosity (n= 1) were bonded using glass plates as restoration to demonstrate that similar layer thicknesses of the resin cements at the two viscosities were obtained through the implemented bonding methodology. After complete resin cement polymerization, the glass plates were detached and the cement layers (at 3 different points) were measured in a contact profilometer (JIS 1994, cut-off= 2.5 mm, range= 800; Mitutoyo SJ 400 Profilometer, Mitutoyo). It was possible to verify similar thicknesses between the resin cement layers, regardless of the resin cement viscosity: mean of 57.05 µm (SD 2.51) for high viscosity, and 55.63 µm (SD 3.14) for low viscosity.

2.4 Shear bond strength under static testing (s-SBS)

As previously mentioned, the shear devices were matched with their corresponding analog metallic pair (Fig. 1E) before the test. Then, the s-SBS (n= 15) was determined in a universal testing machine (Instron 6022; Instron, Norwood, United States) at a crosshead speed of 1 mm/minute. The load was applied with a stainless-steel load applicator (Ø= 5 mm) and 1 KN load cell [26] connected to the test device (Fig. 1F). It is important to note in Fig. 1F that the load applicator acts on the shear device, which in turn acts on the specimen. After each assay, the results at a load of failure by debonding were recorded in Newton and the s-SBS was calculated according to the formula: S = L/A; where "S" is the strength (MPa), "L" is the load required for the failure of the specimen (N), and "A" is the interface area of the specimen, which was individually measured (overall mean = 4.03 mm^2; SD = 0.32 mm^2)

2.5 Shear bond strength under cyclic fatigue (f-SBS)

The samples (n = 15) were tested under cyclic fatigue loading [10,13,18] with the same static shear bond strength device in a dedicated fatigue machine. To do so, an adapted machine with a pneumatic system and a digital display (ACTA, Amsterdam, The Netherlands) was used. Cyclic loading was applied with a hexagon stainless-steel load applicator (Ø= 13 mm) connected to the test device under distilled water (Fig. 1G). The same observation of the static test is made: the load applicator acts on the shear device, which in turn acts on the specimen. The load was initially implemented with 10 N for 10,000 cycles (Fig. 1H) and incremental steps of 5 N for 10,000 cycles were applied until sample failure debonding or the maximum load of the machine – 80 N (survival). The load data (Newton) and the number of cycles for failure were recorded and the fatigue shear was also calculated according to the formula S= L/A, where the area was individually measured (overall mean = 3.92 mm^2; SD = 0.30 mm^2).

2.6 Failure analysis

The fracture pattern was determined under a stereomicroscope (Olympus, Shinjuku, Japan), and classified into two types: 1) adhesive failure at the interfacial region between the resin cement and ceramic/dentin substrate (adhesive areas up to 50 %); 2) cohesive failure in the cement (cohesive areas from 50 %). Representative specimens (adhesive and cohesive) were ultrasonically cleaned (distilled water, 5 min), air-dried, gold-sputtered (Edwards S150B, BOC Edwards, Burgess Hill, United Kingdom), and analyzed under Scanning Electron Microscopy (SEM, Evo LS15, Carl Zeiss, Gottingen, Germany) at 100 × magnification to observe the failure pattern.

2.7 Topographic analysis

Additional dentin and lithium disilicate ceramic specimens (n= 1) with respective surface treatments (acid etching followed by adhesive or silane coating application) were confectioned to be inspected regarding the topographical changes. The specimens were gold-sputtered (Edwards S150B, BOC Edwards) and analyzed by SEM (Evo LS15, Carl Zeiss) at 1,000× and 5,000× magnifications.

2.8 Finite element analysis (FEA)

A three-dimensional finite element analysis (FEA) was used to validate the bond strength methodology introduced herein. An elastic modulus (E) of 10.68 GPa (Variolink II, Ivoclar) [30] and a Poisson ratio (v) of 0.34 [31] for the resin cement and E of 83.5 GPa and v of 0.21 [25] for the lithium disilicate ceramic were determined.

The model was composed of a ceramic substrate and cylinder, a resin cement layer (40 µm), and a steel structure loading consisting of 81.169 parabolic tetrahedron solid elements. The test was carried out using the FEMAP software program (FEMAP 2019.1, Siemens PLM Software, Plano, United States), and the NX Nastran software program (NX Nastran, Siemens PLM Software).

The nodes were fixed outside of the substrate disc, which was bonded to the ceramic cylinder, and in the loading part. The interface surfaces between the bonded ceramic cylinder and the loading part were modeled with contact surfaces with a coefficient of friction of 0.45 and the external surface of the loading part of the steel frame was loaded with the average load resulting from the tests. The maximum tensile stress (solid maximum principal stress) in the resin cement layer using the average value with no corner data was calculated with a load of 80 N. Next, the maximum tensile stress in the resin cement layer was calculated by the linear combination of stresses and shrinkage. A post-gel shrinkage coefficient of the resin cement of 0.005 was used, which is comparable to composites [32,33].

2.9 Data analysis

A statistical software program (IBM SPSS Software, IBM, Armonk, United States) was used to perform the analysis with a significance level of 0.05, discarding the cohesive failures, since they do not represent the real bond strength value. Data were diagnosed as non-normal by the ShapiroWilk test (p= 0.03; p= 0.029; p= 0.152; p= 0.003; p= 0.003; p= 0.444; p= 0.002; p= 0.475) and non-homoscedastic by the Levene test (p= 0.031). A factorial analysis of variance (ANOVA, 2×2) was performed to compare "viscosity * loading mode" for both substrates (with independent comparisons). Due to non-normal data, bootstrapping procedures (1000 resamplings; 95% CI, bias-corrected and accelerated - BCa) were implemented to obtain greater reliability of the results in order to correct deviations from the normality of the sample distribution and to present a 95% CI for differences among means [34]. Furthermore, shear bond strength under fatigue and the number of cycles for failure data were subjected to survival analysis for non-normal data (Kaplan Meier and Log-Rank Mantel-cox post-hoc tests) to access the survival probability through the testing steps. Failure mode, topographic, and FEA analyses were qualitatively evaluated.

3. Results

The mean and standard deviation of s-SBS and f-SBS are presented in Table 3.

By discarding the cohesive failures in the analysis, some cases reduced the group size by 30%. To ensure the representativeness of the data, a power (by mean of averages, OpenEpi) [28] was calculated for the final bond strength data, comparing the high (mean= 31.51, SD= 13.98, sample size= 10.5; considering only adhesive failures) and low group (mean= 19.91, SD= 3.61, sample size= 15; no cohesive failures) of the lithium disilicate substrate and power of 81.82% was achieved.

The two-way ANOVA showed a statistically significant effect for all factors for lithium disilicate substrate: viscosity ($F= 7.322$, $p= 0.010$), loading mode ($F= 34.659$; $p= 0.000$) and the loading mode*viscosity interaction ($F= 8.637$, $p= 0.006$), although the viscosity factor did not show differences among groups ($p= 0.066$ for s-SBS, $p= 0.764$ for f-SBS). Multiple comparisons revealed that groups of different resin cement viscosities behave similarly in each loading mode and that cyclic loading regimen had detrimental effect on the adhesion of both resin cements.

The two-way ANOVA showed a statistically significant effect for loading mode for dentin substrate ($F= 9.125$; $p= 0.005$), although the low viscosity group presented similar results between static and fatigue load ($p= 0.506$). The analysis of variance detected a statistical difference for fatigue load groups ($p= 0.003$) and no statistical difference for the viscosity factor ($F= 1.538$, $p= 0.224$). Finally, the loading mode*viscosity interaction was not statistically different ($F= 2.766$, $p= 0.106$). Multiple comparisons demonstrate that the low and high viscosities behave similarly under static loading, while the low viscosity had the best performance when under cyclic loading. Regarding survival probabilities (Kaplan-Meier and Log-Rank Mantel-cox), the dentin substrate with high viscosity cement presented the worst performance (Fig. 2).

Most failures were adhesive for the lithium disilicate substrate (70–100%), while more cohesive failures occurred for the dentin substrate, mainly in the group with low viscosity and under static loading (50%) (Table 3). There was a clear negative effect on the shear bond strength under cyclic loading, with a 16–56% of bond strength decrease, with special attention to high viscosity, which appears to degrade more than low viscosity in the tested condition (Table 4).

SEM analysis of the tested samples exposed a clear difference between the failure mode (adhesive and cohesive), showing the adhesive area delimited by the tapes (Fig. 3). The topographic analysis revealed a distinct pattern comparing the disilicate and dentin surface, according to the microstructure of each material and the surface treatments performed (Fig. 4). FEA analysis with a load of 80 N demonstrated that there was a stress concentration on the top of the cement layer and that the model deforms when the load is applied (Fig. 5)

4. Discussion

This in vitro study by means of shear bond strength analysis is one of the first to evaluate the shear bond strength under fatigue cyclic loading and to evaluate the survival probabilities of lithium disilicate ceramic bonded to a resin cement and to identify the influence of cyclic loads on the impairment of the adhesive interface. Our findings showed that resin cement viscosities did not influence

the shear bond strength (static and cyclic loading) of lithium disilicate as a substrate. Nonetheless, the high viscosity resin cement impairs the bond strength under cyclic loading when the substrate is dentin. Thus, the first hypothesis that different viscosities of resin cement would induce similar bond strength was partially accepted. In addition, a negative influence of the cyclic loading approach was observed in most evaluated scenarios, except when the low viscosity resin cement is used for the dentin substrate. Therefore, the second hypothesis that fatigue shear would negatively affect the shear bond strength was also partially accepted.

The most striking contribution to emerge from this research is the possibility of predicting survival probabilities from shear data under cyclic loads. A loss of 16–56 % of the bond strength in the bonded ceramic sets is estimated after cyclic loads underwater, although not statistically significant for one group (low-viscosity cement on dentin substrate; Tables 3, 4, and Fig. 2). These findings are in agreement with fatigue mechanism concepts, indicating that any component in normal service that is loaded far below its critical load, either continuously or under repetitive conditions, tends to fail due to fatigue [16,17,21,22]. Fatigue involves a complex degradation phenomenon via slow crack growth (influence of water) or cyclic fatigue mechanisms and can be defined as the degradation (weakening) of a structural component under the influence of mechanical, chemical, biological stress, or a combination of them [16]. In this sense, considering our study, we believe that the adhesive interface and the interaction between two substrates subjected to shear under intermittent cycles also follow the same principles. Furthermore, the applied cyclic fatigue approach in turn enabled application of progressive shear loads with the same protocol for all sets and allowed the analysis of survival rates [10,13,16–18].

A relevant finding of this research is that the resin cement viscosities did not influence the shear bond strength to lithium disilicate substrate and presented similar survival rates (Fig. 2). In a related context, Barbon et al. [9] evaluated the microtensile bond strength between feldspathic glass-ceramic and resin composite with 3 different resin cement viscosities. Even with an inorganic filler ranging from 55 wt% (low viscosity) and 75 wt% (high viscosity), high viscosity was found to be similar to low, which was similar to intermediate viscosity. If we take into account the similar behavior between high and low viscosities, these findings are in line with our study for the ceramic substrate. This fact shows that even large variations in the % weight of fillers, of about 20 % [9], and small variations as in the present study (6 %, Table 1), were not enough to influence the bond strength of ceramics and composite resin substrates. It is likely that all resin agents tested in the study could interpenetrate the surface defects and accommodate some elastic and plastic deformation [9].

On the other hand, a different behavior was observed for the dentin substrate: even if the viscosity had no effect on the bond strength under static mode, the high viscosity after cyclic fatigue loading impairs the bond strength (Table 3) and survival probabilities (Fig. 2). The dentin substrate is known to be critical, humid, porous, and contains a significant amount of minerals within an organic matrix. This heterogeneous structure and surface morphology likely make dentin less inclined to bond with dental adhesives [35,36]. Considering the interfaces involved in this assembly (ceramic – resin

cement and resin cement – dentin), it is expected that a more unstable and less resistant bond will be found in the dental substrate [24]. Moreover, dentin topographic pattern generated by phosphoric acid etching and adhesive coating may compromise the adhesion [37]. The basic bonding mechanism to dentin is essentially an exchange process involving the replacement of minerals removed from the hard dental tissue by resin monomers, which become micro-mechanically interlocked in the created porosities upon setting, in turn creating a hybrid layer [19]. As we can see in Fig. 4, it is possible to note the exposed dentinal tubules. The tubules may not have been completely filled by resin cement, thereby leaving flaws and defects acting as a source for failure initiation sites [19].

Another important observation is regarding the high degradation of the high viscosity resin cement after cyclic loading (shear underwater immersion – up to 30 h), with a bond strength decrease of 56 % for ceramic substrate and 51 % for dentin. Indeed, closer inspection of data (Table 4) shows a trend of viscosity behavior. It seems that low viscosity resin cement exhibits less degradation after intermittent cycles, showing greater stability. This behavior can be explained by some authors who investigated polymer-based materials and found differences among the materials after aging conditions [38]. A strong susceptibility to hygroscopic and hydrolytic effects to varying extents is dependent upon their chemistry and structure [39]. More fillers of the high viscosity cement (+ 8.4 vol%; + 6 wt%) and less monomer content (− 5.9 wt%) can stimulate more swelling processes, plasticization, and thus a higher risk of molecular structure decomposition [38], affecting shear bond strength after cyclic loading.

Although adhesive and mechanical performance in another setup depended on the topography, the type of defects and irregularities after surface treatment, and the ability of the luting agent to fill and seal surface irregularities [7–9,13], the same is valid for the present study. The shear bond strength evaluated is the result of an interaction of all factors involved in the restorative set, such as surface treatments which are capable of generating micromechanical interlocking; the etched ceramic, which should be strengthened by a luting agent that is capable of penetrating surface defects; and substrates (Fig. 4), with dentin being the most critical and challenging component.

Regarding the shear setup, the FEA analysis (Fig. 5) shows the stress concentrated at the top of the layer, with similar characteristics to those found in the knife-edge chisel classical bond strength method [20]. Thus, there are inherent limitations to the shear method, but this does not invalidate the findings, and it helps to enable developing superior materials and new bond strength methodologies [3,4]. To the authors' knowledge, this loading mode has never been evaluated in a bond strength test, and therefore it is now understood that cyclic fatigue loading mode also affects the adhesive interface of the restored sets; in turn, considering the premises involving the fatigue of structures [16,17,40] and the finding of this current study, we emphasize that it plays a significant role in evaluating the bond stability of bonded joints.

Despite these interesting results obtained in this study, there are still some concerns. One of the main limitations is the absence of long-term aging (water storage and thermal cycling) and the absence of resin cement viscosity evaluation. Future studies should consider thermocycling and water storage

aging conditions to challenge the assemblies [19,23] and also a viscosity assessment. Furthermore, our study considered standardized thicknesses for both resin cement viscosities. This measure allows to eliminate a possible confounding bias of the results; however, it is clinically expected that low viscosity resin cements generate a thinner layer of the luting agent [14,41]. Therefore, the thickness variable should be evaluated in future studies. Another limitation is regarding the fatigue shear machine equipment, which is not commercially available, and therefore special attention should be given to methodological reproducibility. Even so, our study is relevant to demonstrate the detrimental effect of cyclic loading in shear inducing mechanical fatigue on bonded joints in Dentistry.

5. Conclusion

- Cyclic loading shows deleterious effects on the adhesive behavior and survival probabilities of bonded lithium disilicate restorative assemblies, regardless of resin cement viscosity (high or low).
- In contrast, the different viscosities of resin cements influence the shear bond strength and survival rates of dentin substrate submitted to cyclic loading mode in which a low viscosity induces better performance.
- Taking into account that lithium disilicate restorations sets are subjected to challenges in the oral cavity, the bond strength under cyclic loading regime outcomes seems to be more reliable than a static one.

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TABLES

Table 1. Description of materials, commercial name, manufacturer, composition, filler % weight, filler % volume, and batch number.

Material	Commercial name/manufacturer	Composition*	Filler % wt.	Filler % vol.	Batch number
5% Hydrofluoric acid	IPS Ceramic Etching Gel/ Ivoclar, Schaan, Liechtenstein	< 5% hydrofluoric acid	-		Y48112
35% Phosphoric acid	K-etchant syringe gel/ Kuraray Noritake Dental Inc., Okayama, Japan	Aqueous solution of phosphoric acid and colloidal silica			BV0048
Dual-curing resin cement	Variolink N High Viscosity (Catalyst)/ Ivoclar	Barium glass filler and mixed oxide, dimethacrylates, ytterbium trifluoride, initiators and stabilizers, and pigments	77.2 %	52.0 %	YZ1262
	Variolink N Low Viscosity (Catalyst)/ Ivoclar	Barium glass filler and mixed oxide, dimethacrylates, ytterbium trifluoride, initiators and stabilizers, and pigments	71.2 %	43.6%	YZ1263
	Variolink N Base/ Ivoclar	Barium glass filler and mixed oxide, dimethacrylates, ytterbium trifluoride, initiators and stabilizers, and pigments	73.4 %	46.7 %	YZ1282
Dual-curing total-etch adhesive	ExciTE F DSC/ Ivoclar	Phosphonic acid acrylate, dimethacrylates, hydroxyethyl methacrylate, highly dispersed silicon dioxide, ethanol, catalysts, stabilizers, and fluoride	-		Y50449
Lithium disilicate glass-ceramic	IPS e.max CAD, HT A2, C14/ Ivoclar	SiO_2, Li_2O, K_2O, P_2O_5, ZrO_2, ZnO, other and coloring oxides			Y52153
Silane-based coupling agent	Monobond N/ Ivoclar	Alcohol solution of silane methacrylate, phosphoric acid methacrylate, and sulphide methacrylate			Y45831

*The chemical composition is described according to the manufacturers' information.

Table 2. Experimental design (n= 15).

Ceramic	Study´s factor		
	Resin cement viscosity (2 levels)	Loading mode (2 levels)	Substrate (2 levels)
Lithium disilicate	High Low	Static (s-SBS) Cyclic fatigue (f-SBS)	Dentin Lithium disilicate

Table 3. Mean and standard deviation (SD) of the shear bond strength under static (s-SBS) and cyclic fatigue loading (f-SBS) in MPa of lithium disilicate bonded to dentin and lithium disilicate substrates[*].

Resin cement	Lithium disilicate substrate						Dentin substrate					
	s-SBS			f-SBS			s-SBS			f-SBS		
	Mean (SD)	Failure types		Mean (SD)	Failure types**		Mean (SD)	Failure types		Mean (SD)	Failure types	
		Adhesive	Cohesive		Adhesive	Cohesive		Adhesive	Cohesive		Adhesive	Cohesive
High viscosity	31.51 (13.98) ^{Aa}	70%	30%	13.37 (4.47) ^{Ba}	86%	14%	14.15 (5.83) ^{Aa}	80%	20%	6.89 (3.28) ^{Bb}	87%	13%
Low viscosity	19.91 (3.61) ^{Aa}	100%	-	13.85 (3.29) ^{Ba}	86%	14%	13.50 (6.30) ^{Aa}	50%	50%	11.39 (3.45) ^{Aa}	73%	27%

* Two-way ANOVA adjusted by bootstrapping. Statistical differences are represented by lowercase letters in the column (compare viscosity) and uppercase letters in the row (compare loading mode) within each substrate.
** Survival: One specimen of each resin cement of lithium disilicate and fatigue shear survived the test.

Table 4. Decrease percentage of bond strength after cyclic fatigue tests (f-SBS).

Resin cement	Lithium disilicate substrate	Dentin substrate
High viscosity	56%	51%
Low viscosity	30%	16%

FIGURES

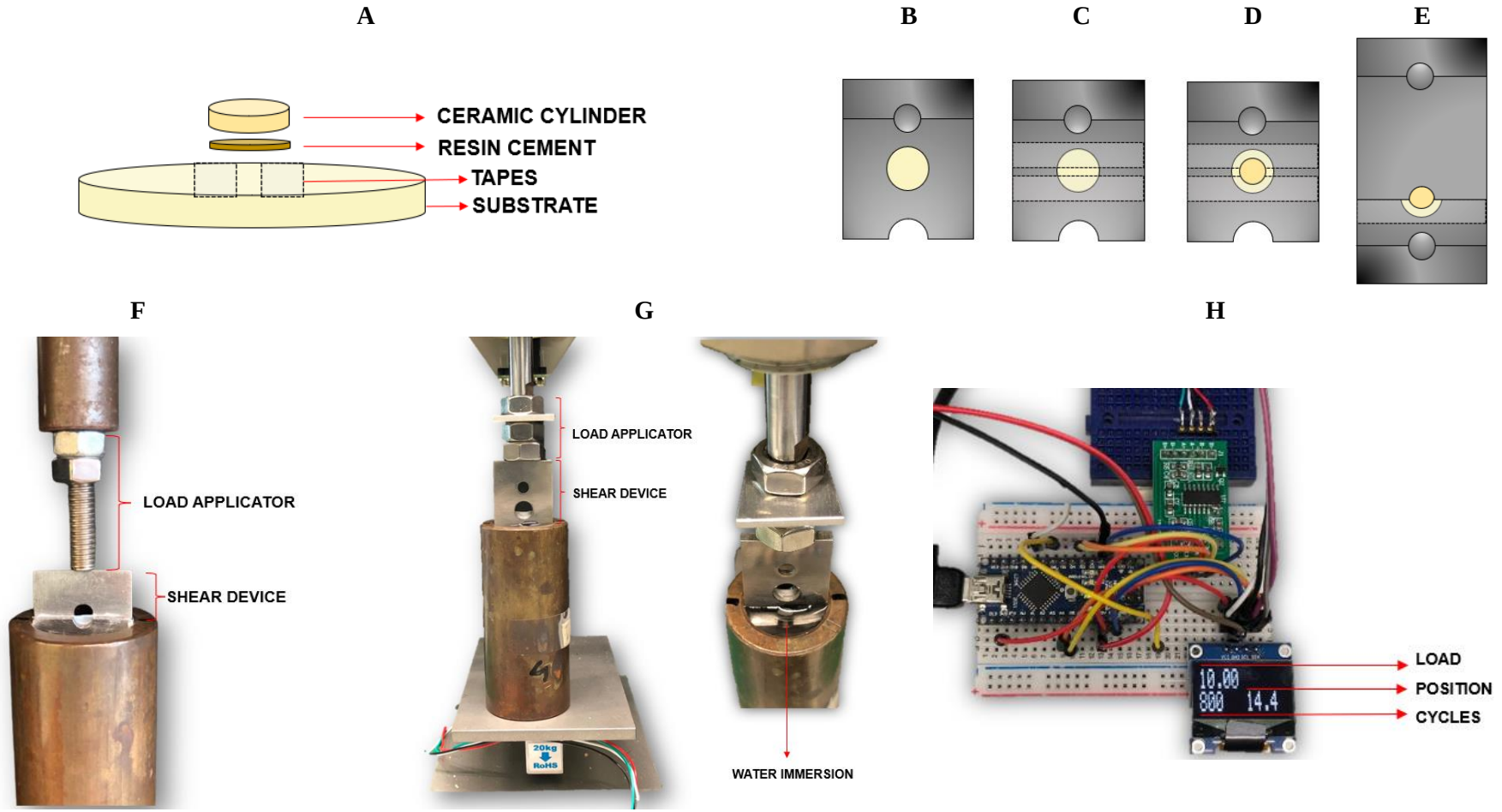
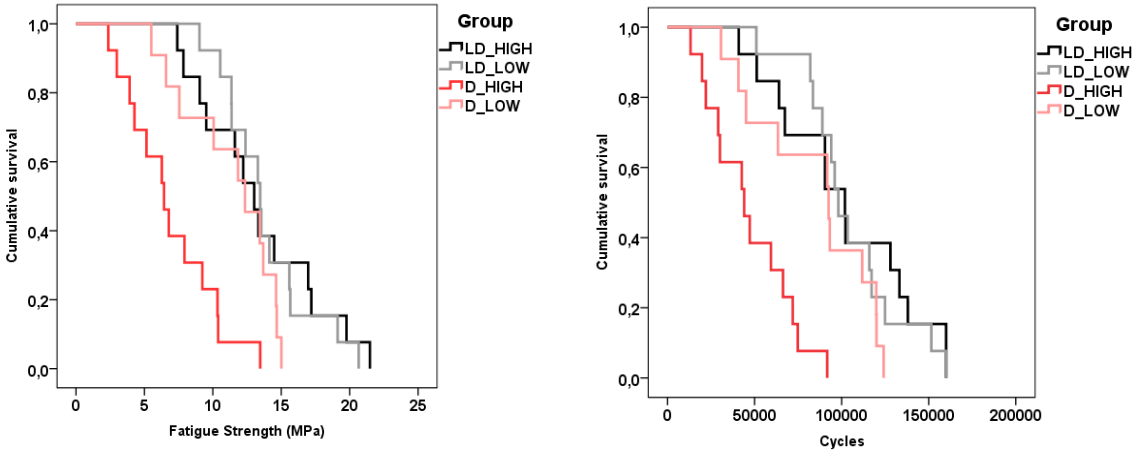
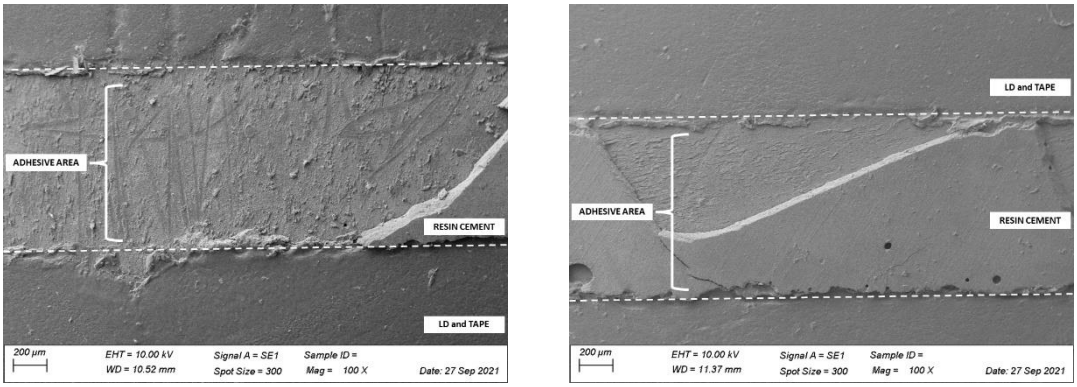


Figure 1. Schematic illustration of the restorative assembly, shear device, and shear setup test (static and fatigue). A. Restorative set (three-layer assembly); B. Substrate positioned on the shear device; C. Tapes positioned to delimit the adhesive area; D. Ceramic cylinder positioned over resin cement and substrate; E. Pair-fitted shear devices; F. Static shear setup positioned under the load applicator (Note: The applicator acts on the shear device, which in turn acts on the specimen.); G. Fatigue shear setup device (front and top view) positioned under the load applicator and the restorative set under distilled water immersion (Note: The adhesive area is perpendicular to the load application); H. Machine screen showing test parameters (load, the position of the load applicator in relation to the load cell, and cycles).



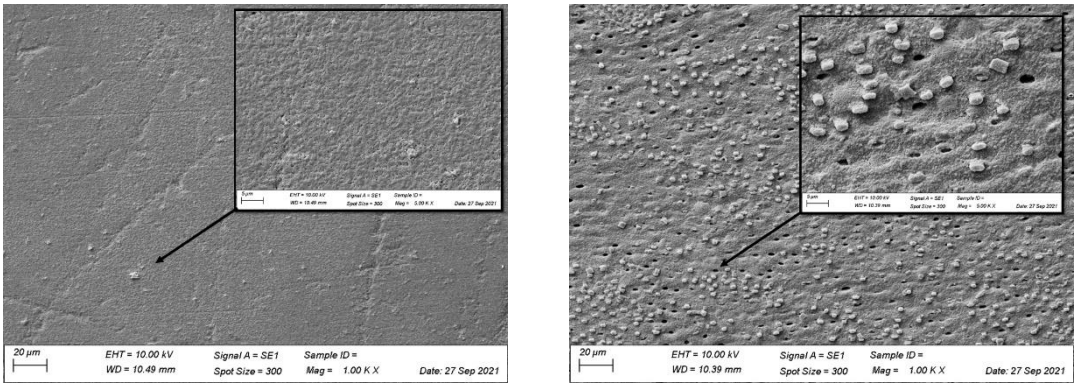
A **B**

Figure 2. Survival graphs obtained by the Kaplan-Meier and Mantel-Cox (log-rank) test: 'A' for fatigue strength in MPa and 'B' for the number of cycles for failure. LD: Lithium disilicate substrate; D: Dentin substrate.



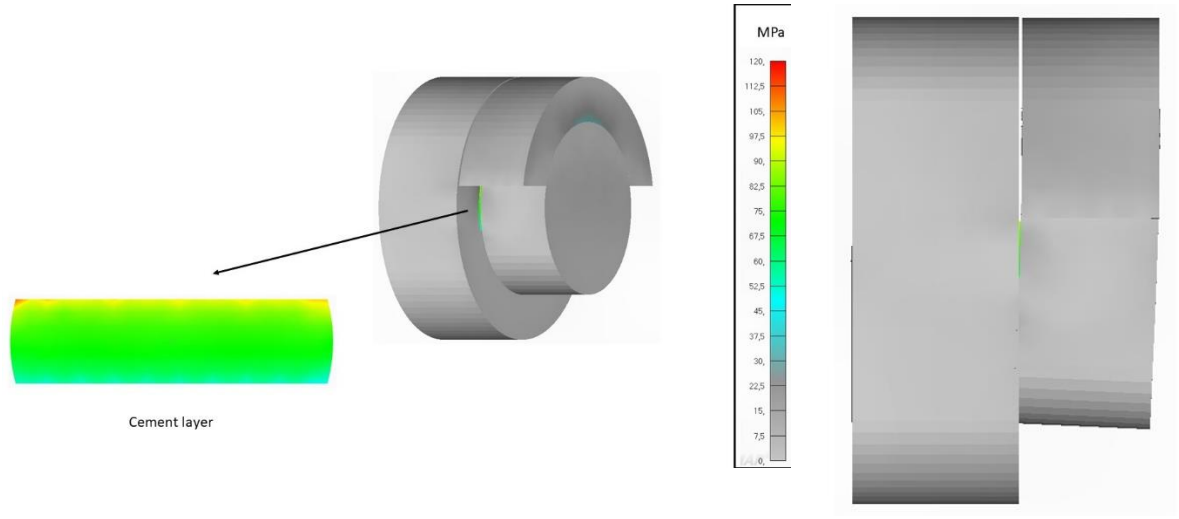
Adhesive failure **Cohesive failure**

Figure 3. SEM micrographs (100×) demonstrating the pattern of failure mode. It is possible to observe the adhesive area restricted to positioned tapes (parallel dotted lines). In addition, the resin cement layer is visualized, more attenuated in the figure on the right, which characterizes the type of cohesive failure.



Lithium disilicate substrate **Dentin substrate**

Figure 4. Topographic images (1000×; 5000×) obtained by SEM show the HF etched and silane coated disilicate surface as smoother and more homogeneous (even at higher magnification); on the dentin surface, it is possible to observe both the dentinal tubules after the phosphoric acid etching and the particles of the applied adhesive.



A **B**

Figure 5. FEA model with 40 µm of cement layer thickness and application of 80 N (maximum load). A. Tensile stress distribution highlighted throughout the cement layer with stress peak at the top of the cement layer and lower intensity on the face opposite to the application of the load; B. The side view presents deformation of the ceramic cylinder subjected to load application and consequent higher tensile stress on top of the cement layer.

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5 ARTIGO 4: Do resin cement viscosity and ceramic surface etching influence the fatigue performance of bonded lithium disilicate glass-ceramic crowns?

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Do resin cement viscosity and ceramic surface etching influence the fatigue performance of bonded lithium disilicate glass-ceramic crowns?

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Running title: Effects of resin cement viscosity on the fatigue of bonded crowns

Abstract

Objective. To assess the effects of a resin cement in high and low viscosity and distinct conditioning of the intaglio surface of lithium disilicate glass-ceramic crowns on fatigue performance of the crowns.

Methods. Prosthetic preparations (full-crown) in resin epoxy and crowns in lithium disilicate glass-ceramic were machined and allocated considering 2 factors (n=10): "surface treatment" (HF – 5% hydrofluoric acid etching, followed by silane application; or E&P–self-etching ceramic primer) and "resin cement" (high or low viscosity). The preparations were etched with 10% hydrofluoric acid and an adhesive was applied. The intaglio surfaces of the ceramic crowns were treated as aforementioned (HF or E&P) and luted with high or low viscosity. The bonded sets were subjected to fatigue testing (step-stress approach: initial load of 200N, step-size of 50N, 10,000 cycles/step, 20 Hz) and complementary analyses (fractographic, topographic, and cross-sectional bonded interfacial zone analyses) were performed.

Results. Treatment with HF and silane with high viscosity resin cement (955N/156,000 cycles) and E&P with low viscosity resin cement (1090N/183,000 cycles) showed the best fatigue performance (statistical similarity between them). The failures originated from defects of the cement-ceramic interface, and the HF treatment induced a more pronounced topographical alteration.

Significance. Distinct topographical patterns from the HF and E&P treatments induced better fatigue results for the specific viscosity of the resin cement. Therefore, the fatigue performance depended on the existing topography, type of intaglio surface's defects/irregularities after surface treatment, and how the luting agent filled the irregularities.

Keywords: Cementation. Ceramics. Computer-Aided Design. Dental bonding. Dental cements. Dental Porcelain.

Highlights

- Fatigue behavior of lithium disilicate crowns depends on the cement viscosity and the topographical changes resulting from ceramic surface treatment.
- Superior fatigue failure results are obtained with HF/silane + High viscosity; or Etch & Prime + Low viscosity.
- The defects at the ceramic intaglio surface need to be filled by the luting agent for fatigue improvements.
- The luting agent viscosity play a crucial role for filling the defects.
- HF etching induced a more pronounced topographical alteration compared to the Etch & Prime agent.

1. Introduction

Lithium disilicate glass-ceramic posterior crowns fashioned by computer-aided design and manufacturing (CAD/CAM) exhibit a satisfying survival rate of 83.5% after functioning for 10 years [1]; and they are considered to be fracture-load-resistant restorative material that provide reliable clinical performance [2] and excellent esthetic results [3]. In fact, the microstructure (glass matrix and interlocking lithia crystals) and processing of lithium disilicate ceramic allow a highlighted balance between the optical and mechanical properties [2].

Resin bonding is the gold standard for retention and strengthening of lithium disilicate restorations, but it requires multiple pretreatment steps of the bonding surfaces [4-7]. Lithium disilicate is an acid-sensitive ceramic due to the glass matrix and requires accurate surface etching in order to achieve stable adhesion to the substrate, due to the micromechanical and chemical bonding mechanisms [8-10]. The micromechanical interlocking between ceramic and resin cement at the intaglio surface is based on the creation of surface micro irregularities and higher roughness promoted by acid etching [2]. For this, hydrofluoric (HF) acid, followed by application of a silane coupling agent, is the classic and well-established procedure for glass-ceramics [9,10]. However, studies have already reported self-etching ceramic primer to be a single step alternative for surface treatment that decreases the hazardous effects of toxicity compared to HF acid [8,11].

For restorations machined by CAD/CAM systems, it is important to note that processing modifies the surface micromorphology and introduces defects at the surface and subsurface of the intaglio surface of the ceramic restorations [12-14]. Subsequently, the surface still interacts with chemical agents for the surface conditioning (for instance, HF acid) performed prior to bonding, triggering surface topographical changes [13,15]. Subsequently, there is the interaction of the treated ceramic surface with the resin cement used from bonding the restoration on the substrate, with the expectation that the cement fills the surface defects [8,11,16].

Resin cements consist of inorganic fillers embedded in a polymer matrix [10]; they present different characteristics according to the resin monomer (matrix), filler (inorganic particles), and coupling agents [17]. Among resin cement properties, viscosity is a measure of a liquid's resistance to forces that tend to cause it to flow [18]. In resin cements, it depends on the composition and ratio of the resin matrix; the content, shape, size distribution, and silane treatment of the inorganic filler interlocking between filler particles; and interfacial interaction between filler particles and the matrix resin [19-22].

Viscosity, as a rheological property, is a key parameter that influences perceived differences in handling behavior of resin composites [21]. Based on this assumption, the viscosity of resin cements can change the filling potential of defects resulting from machining and surface conditioning and, consequently, influence strengthening of the restoration set through better dissipation of mechanical stresses [4,23]. Understanding the effects of the viscosity and the capacity of resin cement to completely fill defects introduced on the intaglio surface of the ceramic material is essential for predicting the mechanical performance of the restorative system. In this sense, the strengthening mechanism proposed

suggests creation of a “resin-ceramic hybrid layer,” and the stressing patterns near the region of a critical defect become sensitive to the characteristics of this layer [23].

However, the influence of resin cement viscosity and proper interpenetration of the etched surface on the performance of restorative systems under intermittent cyclical loading (mechanical fatigue) have not been completely clarified in the literature. Therefore, this study aimed to answer a main question: Does the viscosity of resin cement and its potential for filling the irregularities/defects created by distinct ceramic surface etchings influence the fatigue performance of lithium disilicate glass-ceramic crowns? Based on this, the objective of this study was to evaluate a resin cement in high and low viscosity and distinct surface ceramic treatments (HF acid plus silane and self-etching ceramic primer) on the fatigue performance of bonded machined lithium disilicate crowns. The assumed null hypotheses were: (1) the surface etchings will induce similar fatigue behavior; (2) resin cement viscosities will not affect the fatigue behavior.

2. Material and methods

2.1. Materials and study design

The general description and characteristics of the materials used are described in Table 1.

This study was designed in 4 experimental groups, considering 2 factors (n=10, Table 2):

i) “Surface treatment” of the intaglio surface of the lithium disilicate crown: hydrofluoric acid + silane coupling agent (HF groups) and self-etching ceramic primer (E&P groups).

ii) “Resin cement” in different viscosities: high and low.

2.2. Specimen preparation

Simplified prosthetic preparations simulating a posterior single full-crown (molar, N=40) were made of an epoxy resin-reinforced glass-fiber material (Protec Produtos Técnicos Ltda., São Paulo, SP, Brazil) and utilized as the substrate (Kelly et al., 2010). The preparations were machined from 11 mm diameter rods of an epoxy-glass cloth in a mechanical lathe (Diplomat 3001, Nardini, Americana, SP, Brazil), with all identical preparations (5.32 mm high; internal angle radii of 0.5 mm; axial walls with 8º inclination; total occlusal convergence angle of 16º with rounded corners). Next, a randomly selected preparation was scanned (inEos Blue, Dentsply Sirona Dental Systems GmbH, Bensheim, HE, Germany); the image transferred to the software (CEREC in-Lab 3D, v4.1, Dentsply, Sirona Dental Systems GmbH); and a simplified molar monolithic crown designed, considering a cementation space of 80 µm and 1.0 mm of final occlusal and axial thickness.

Then, 40 blocks of lithium disilicate (IPS e.max CAD, Ivoclar-Vivadent, Schaan, Liechtenstein) were machined (CEREC inLab MC XL, Dentsply Sirona Dental Systems GmbH) with diamond rotary instruments (step bur 12S a cylinder-pointed bur 12S) under water-cooling with a lubricant into identical simplified single crowns, simulating a molar. The sprue cutting was performed using a diamond disc (American Burrs, Palhoça, SC, Brazil) and border adjustments/finishing were made with siliconized

stones and rubbers (American Burrs). Next, the lithium disilicate crowns were crystallized according to the manufacturer's instructions (840°C, 7 min vacuum, Vacumat 6000 MP, VITA Zahnfabrik, Bad Säckingen, BW, Germany) using a firing paste (IPS Object Fix Flow, Ivoclar-Vivadent). Then, the crowns (N=40) were randomly allocated (https://www.randomizer.org) according to groups (n=10) (Table 2).

2.3. Bonding procedures

Each set (preparation and crown) was checked for internal adaptation and cleaned (isopropyl alcohol, 5 min) in an ultrasonic bath (1440 D, Odontobras Medical and Dental Equipment, Ltda., Ribeirão Preto, SP, Brazil). The preparations were etched with 10% hydrofluoric acid (Condac Porcelana, FGM Dental Group, Joinville, SC, Brazil) for 60 sec; cleaned in an ultrasonic bath (distilled water, 5 min); air dried; and an adhesive (ExciTE F DSC, Ivoclar-Vivadent - Variolink system) was applied for 10 sec, according to the manufacturer's recommendation.

The intaglio surfaces of the crowns were treated according to "surface treatments". The samples from the HF groups were etched with 5% hydrofluoric acid (IPS Ceramic Etching Gel, Ivoclar-Vivadent) for 20 sec, air-water sprayed for 30 sec, and air-dried for 30 sec. Next, silane coupling agent was applied (Monobond N, Ivoclar-Vivadent), which was then actively scrubbed for 15 sec and allowed to react for another 45 sec, with posterior air-dry for 30 sec. The ceramic surfaces of the E&P groups were treated with the self-etching ceramic primer (Monobond Etch & Prime, Ivoclar-Vivadent) using a microbrush for 20 sec actively, left to react for 40 sec, air-water sprayed for 30 sec, and air-dried for 30 sec, featuring a simplified conditioning system. All crowns were subjected to an ultrasonic bath (distilled water up to 5 min) after the ceramic surface treatments (i.e., after HF and one-step E&P etchings). Then, the dual-curing resin cement (Variolink N, Ivoclar-Vivadent) in high or low viscosity (catalyst) and base were mixed at a 1:1 ratio with the same base and applied on the intaglio surface of the crowns. Each crown was positioned on a preparation, and the assembly was seated under a static load of 7.5 N. After removing the excess resin cement, the set was photo-activated for 20 sec on each side of the specimen (0º, 90º, 180º, 270º, and on the top surface). All samples were stored (37°C, distilled water) for 24 hours – 7 days until the fatigue tests.

2.4. Fatigue test

The sets were submitted to a fatigue test in an electronic testing machine (Instron ElectroPuls E3000; Instron, Norwood, MA, USA) using the step-stress methodology. Load pulses were applied by stainless steel ball piston (Ø=40 mm) at a frequency of 20 Hz [24] with the specimen immersed in distilled water. An adhesive tape (0.1-mm thick) was placed on the occlusal surface of the cemented crown to reduce contact stress concentration, improving contact between the piston and specimen [25].

An initial load of 200N (5,000 cycles to seat the specimen and adjust the piston/specimen contact), followed by progressive load levels of 50N (250N, 300N, 350N, 400N, up to failure detection),

were applied for 10,000 cycles each. The specimens were checked for failures at the end of each loading step under transillumination [26] and the test was terminated if a failure, considered a radial crack, was detected.

2.5. Complementary analyses

2.5.1. Fractographic, topographic, and cross-sectional bonded interfacial zone analyses (Stereomicroscope and Field Emission Scanning Electron Microscopy)

For fractographic analysis, all the specimens were inspected under a stereomicroscope (Discovery V20, Carl-Zeiss, Göttingen, NI, Germany) to determine the region of fracture. Representative failures of each tested group (n=1) were selected, and crown fragments were detached to access the origin of the defect and fractographic characteristics. The samples were gold-sputtered (Denton Desk II, Dentin Vacuum, LLC, Moorestown, NJ, USA) and analyzed by Scanning Electron Microscopy (SEM, JSM-6360, JEOL, Tokyo, Japan) at 22× and 100× magnification.

To inspect topographic changes and microstructure features after surface treatments, the axial walls from a non-bonded additional crown (n=1) were removed using a cutting machine (Isomet 1000, Buehler, Lake Bluff, IL, USA) and a diamond disc, until the complete exposure of the internal occlusal surface, which remained untouched with a disc-shaped format. Then, these surfaces were etched as aforementioned (Table 2), keeping one piece as-machined (to compare surface alterations between milled vs. etched). The milled or etched intaglio surfaces were analyzed by Field Emission Scanning Electron Microscopy (FE-SEM, Sigma 300 VP, Carl Zeiss Ltd., Cambridge, Cambs, United Kingdom) at 250× and 1,000× magnification.

For cross-sectional bonded interfacial zone analysis [5], one luted crown of each tested group (n=1) was randomly selected and sectioned into two halves (Isomet 1000, Buehler Ltd.) to expose the cross-sectional surface (including the ceramic-resin cement-substrate interfaces). The cross-sectional surface was mirror polished (#1200- and #2400-grit silicon carbide paper) in a polishing machine (EcoMet/AutoMet 250, Buehler Ltd.) under water. The bonded zone was inspected for morphology, cement/surface relationship, introduction of the defects to etching, and filling with resin cement (FE-SEM, Sigma 300 VP, Carl Zeiss Ltd.) at 250× and 500× magnification.

2.6. Data analyses

Normal and homoscedastic data distribution (Shapiro Wilk and Levene tests) were detected for fatigue data; they were then subjected to two-way analysis of variance (ANOVA) and the post-hoc Tukey test (Statistix 8.0, Analytical Software, Tallahassee, FL, USA) to evaluate the influence of independent variables (surface treatment and resin cement) on their outcomes. Fatigue data were also subjected to survival analysis (Kaplan Meier and Mantel-cox post-hoc tests) to access the fatigue failure load, cycles for failure, and survival rates (IBM SPSS Software, IBM, Armonk, NY, USA). All statistical

analyses were performed with a significance level of 0.05 and the complementary analyses were qualitatively evaluated.

3. Results

An open-source calculator (www.openepi.com) was used to determine the power of this study by the difference in means of the fatigue data, considering the HF_LOW and E&P_LOW groups. The achieved power was 93.68% by the normal approximation method.

Two-way ANOVA revealed that "surface treatment" ($p=0.0641$, $F=3.65$) and "resin cement" ($p=0.4673$, $F=0.54$) did not have significant influences on the outcome (fatigue failure load and cycles for failure). However, interaction between the independent factors "surface treatments" and "resin cement" was statistically significant ($p=0.0013$, $F=12.10$).

In terms of fatigue findings (Table 3), hydrofluoric acid plus silane combined with high viscosity resin cement (HF_HIGH) and self-etching ceramic primer with low viscosity resin cement (E&P_LOW) performed better statistically. The survival probabilities (Table 4) corroborate those fatigue findings; i.e., HF_HIGH and E&P_LOW groups had higher survival rates compared to the HF_LOW and E&P_ HIGH, in which earlier failures occurred.

Regarding complementary analyses, fractographic analysis showed that the failures (radial cracks) originated from the defects at the cement-ceramic interface (Figures 1 and 2). In the topographic analysis, distinct topographic patterns were observed after surface treatment compared to the as-machined; and they demonstrated the potential of treatments to promote surface alterations (glassy matrix removal and pull out of lithium disilicate crystals) at different intensities being more pronounced with HF etching (Figure 3). In addition, the cross-sectional bonded interfacial zone analysis clearly showed the interface among the substrate-resin, cement-ceramic, and distinct topographic patterns introduced by the surface treatments, revealing unfilled areas of resin cement especially in HF_LOW and E&P_HIGH (Figure 4).

4. Discussion

This study provides further evidence regarding the effect of the different resin cement viscosities and distinct surface treatments on the fatigue performance of machined and bonded lithium disilicate crowns. If HF acid plus silanization with a high viscosity cement and self-etching ceramic primer with a low viscosity cement had the best fatigue performance, then the hypotheses had to be rejected, since distinct fatigue behaviors were found. This means that distinct topographical patterns from the surface etchings induced higher fatigue results when using a resin cement with a specific viscosity. Therefore, for this test scenario, the fatigue performance of the crowns depended on the existing topography, type of interfacial defect after surface treatment, and how the luting agent filled these defects.

It is known that the CAD/CAM machining process influences and introduces surface defects in lithium disilicate ceramic restorations [13-15]. After machining, the most important aspect is that the intaglio surface of these restorations is subjected to surface treatments (HF acid or self-etching ceramic primer) that change the initial topography generated by CAD/CAM and define the final surface characteristics, as demonstrated herein (Figure 3) and through other studies [13,15]. From this viewpoint, the resin cement must be able to fill the surface irregularities (plenty of complexity) after machining/etching to mechanically behavior enhance the restorative system [16,27]. When silane and resin cement seal surface microcracks, they prevent the cracks from opening freely due to crack bridging [28] and they perform a continuous and homogenous interphase (ceramic - resin cement - substrate), thus reducing the flaw population and increasing the energy required for the fracture to spread [5]. However, when air bubbles, reminiscent defects, and areas not completely filled-up by resin cement remain, failure of the restorative set occurs due to accumulation of tensile stress around these critical flaws that exceed the nominal strength of the material (Figures 2 and 4) [29].

As the resulting features of the intaglio surface are defined by the conditioning approach, it is important to pay attention to the shape and size of the defects to understand the relationship between the resin cement's viscosity and micromechanical interlocking [5,30]. Surface treatment with hydrofluoric acid resulted in a more irregular topographic pattern promoted by dissolution of the glass matrix, presenting larger, deeper pores and pulled-out lithium disilicate crystals. The self-etching ceramic primer showed less pronounced surface alterations, with partial dissolution of the vitreous matrix and less pores (Figure 3) [8,11,13-15,30]. In this context, it is possible to note that high viscosity resin cement filled better, deeper, and more acute defects (HF), while a resin cement with a lower viscosity had a higher potential for filling the softer defects (self-etching ceramic primer, Figure 4), which generated a clear consequence on the fatigue behavior of the ceramic restoration (Tables 3 and 4).

In relation to the inherent properties of resin cements, the size and content of fillers play an important role in defining their mechanical behavior [31]. The viscosity increased as the percentage of filler volume was increased [19,32]. In fact, to change the resin cement viscosity used in the present study, the manufacturer modified the percentage by weight of fillers, organic matrix, initiators, and stabilizers of the catalyst pastes, while the base pastes remained the same. Regarding fillers and mixed oxide, 52.2% was used for high and 46.2% for low viscosity. For the organic matrix, 22.0% was used for high and 27.9% for low viscosity, and a slight difference was used for the initiators and stabilizers—0.8% for high and 0.9% for low viscosity. Based on that, it is important to consider the effect of distinct compositions may have presented on the differences observed. Future studies may consider the combined effect of distinct compositions and distinct viscosities to the filling potential and mechanical properties of etched ceramics.

Although the viscosity and penetration potential of resin cements are a critical parameter, other mechanical properties, such as the elastic modulus, are also important for the performance and longevity of these materials, since the elastic modulus is related to cement deformation and the creation of

marginal gaps [31,33]. Spazzin et al. [5] indicated that by increasing the elastic modulus of the luting agent, the magnitude of the stresses is more concentrated at the luting agent layer, reducing the magnitude of stresses that affect the restorative assembly. In this scenario, not only the load required for fracture increased, but also the stresses navigating toward the ceramic have increased difficulty in finding flaws to initiate failure. Moreover, the luting agents may have accommodated some elastic and plastic deformation, which also contributed to the reinforcement effect [5]. However, as discussed, more viscous luting agent may potentially differ to generate intimacy between the ceramic surface defects and infiltrating material, and if the defects (unfilled zones) remain located in the stress concentration area, a premature failure can be triggered.

Despite its fortitudes, this *in vitro* study has inherent limitations, which imply that extrapolation and external validity must be carefully pondered. Among the main limitations are the application of only axial loads (without sliding), the absence of aging, the use of simplified ceramic crowns and preparations, and the use of a resin epoxy material in lieu of a tooth substrate. On the other hand, many of these limitations favor standardization and control of the variables studied, thus allowing for more predictable and reproducible results. Nonetheless, the results of the present study are important for clarifying that with the use of HF acid in lithium disilicate; larger defects are produced and a higher viscosity cement can increase performance of the restorative system. Meanwhile, when using a self-etching ceramic primer, the resulting surface has fewer defects and is optimized with a more fluid resin cement. Therefore, the clinical choice must consider the existing surface topography (irregularities and defects) and the best luting agent option to fill these defects.

5. Conclusion

- Distinct topographical patterns from the HF and E&P etchings induced fatigue improvements when combined with a specific viscosity of the resin cement. Therefore, fatigue performance depends on the topography, features of the intaglio surface's irregularities/defects after surface treatment, and how the luting agent interacts/fills such defects.
- Bonding strategies that combine HF acid and silane with a high viscosity cement or self-etching ceramic primer with a low viscosity cement promoted superior fatigue performance of the bonded machined lithium disilicate glass-ceramic crowns.

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TABLES

Table 1. Description of materials, commercial name, manufacturer, composition and batch number.

Material	Trade market and manufacturer	Main composition*	Batch number
5% HF	IPS Ceramic Etching Gel, Ivoclar-Vivadent, Schaan, Liechtenstein	< 5% Hydrofluoric acid	W14921
10% HF	Condac Porcelana, FGM, Joinville, Brazil	< 10% Hydrofluoric acid	W010819
Dual-curing resin cement	Variolink N High Viscosity (Catalyst), Ivoclar-Vivadent	Barium glass filler and mixed oxide (52.2%**), dimethacrylates (22.0%), ytterbiumtrifluoride (25%), initiators and stabilizers (0.8%) and pigments (< 0.1%)	Y15347
	Variolink N Low Viscosity (Catalyst), Ivoclar-Vivadent	Barium glass filler and mixed oxide (46.2%), dimethacrylates (27.9%), ytterbiumtrifluoride (25%), initiators and stabilizers (0.9%) and pigments (< 0.1%)	Y06775
	Variolink N Base, Ivoclar-Vivadent	Barium glass filler and mixed oxide (48.4%), dimethacrylates (26.3%), ytterbiumtrifluoride (25%), initiators and stabilizers (0.3%) and pigments (< 0.1%)	Y06775
Dual-curing total-etch adhesive	ExciTE F DSC, Ivoclar-Vivadent	Phosphonic acid acrylate, dimethacrylates, hydroxyethyl methacrylate, highly dispersed silicon dioxide, ethanol, catalysts, stabilizers and fluoride	X25002
Firing paste	IPS Object Fix Flow, Ivoclar-Vivadent	Oxides, water and thickening agent	Y36206
Lithium disilicate ceramic	IPS e.max CAD, Ivoclar-Vivadent	SiO_2, Li_2O, K_2O, P_2O_5, ZrO_2, ZnO, Al_2O_3, MgO and colouring oxides	W89815
Self-etching ceramic primer	Monobond Etch & Prime, Ivoclar-Vivadent	Ammonium polyfluoride, silane system based on trimethoxypropyl methacrylate, alcohols, water and colorant	Y27773
Silane coupling agent	Monobond N, Ivoclar-Vivadent	Alcohol solution of silane methacrylate, phosphoric acid methacrylate and sulfide methacrylate	Y19262

* The chemical composition is described according to the manufacturers' information;
** % in weight.

Table 2. Experimental design.

Groups' Codes	Surface Treatment	Resin Cement
HF_HIGH	5% Hydrofluoric acid + silane coupling agent (HF)	High viscosity
HF_LOW		Low viscosity
E&P_HIGH	Self-etching ceramic primer (Monobond Etch & Prime, E&P)	High viscosity
E&P_LOW		Low viscosity

Table 3. Mean of the fatigue test for fatigue failure load (FFL) and cycles for failure (CFF) with 95% confidence intervals.

Group	FFL (N)	CFF (Cycles)
HF_HIGH	955 (856.51-1053.49) [ab]	156,000 (136,302-175,698) [ab]
HF_LOW	815 (751.24-878.76) [c]	128,000 (115,247-140,753) [c]
E&P_HIGH	875 (794.65-955.35) [bc]	140,000 (123,930-156,070) [bc]
E&P_LOW	1090 (949.27-1230.73) [a]	183,000 (154,854-211,146) [a]

* Same lowercase letters on each column indicate statistical similarity depicted by Kaplan Meier and Mantel-Cox post-hoc test.

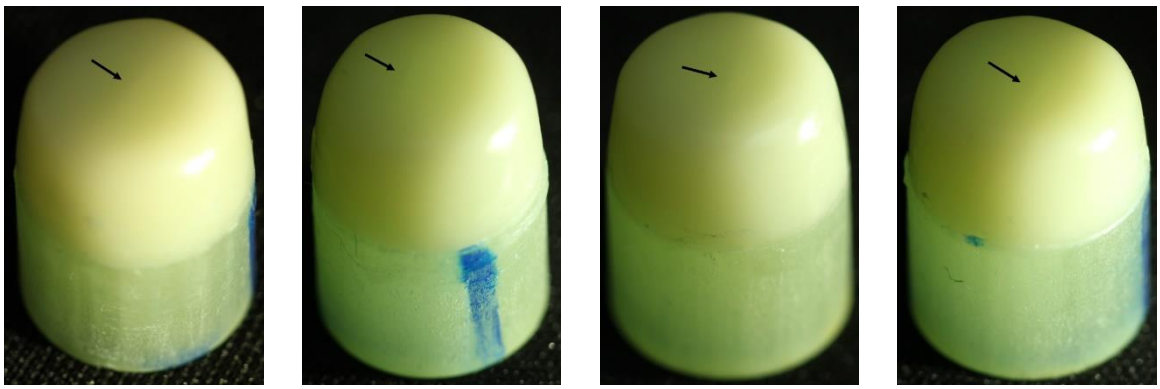
Table 4. Survival rates* of the fatigue test – probability of specimens to exceed the respective fatigue failure load (FFL) and number of cycles for failure (CFF) step without failure (radial crack), and its respective standard error values.

Group	FFL (N) / CFF														
	…	650/ 95,000	700/ 105,000	750/ 115,000	800/ 125,000	850/ 135,000	900/ 145,000	950/ 155,000	1000/ 165,000	1050/ 175,000	1100/ 185,000	1150/ 195,000	1200/ 205,000	1250/ 215,000	1300/ 225,000	1350/ 235,000
HF_HIGH	1	1	0.90 (0.09)	0.80 (0.13)	0.80 (0.13)	0.60 (0.15)	0.60 (0.15)	0.60 (0.15)	0.40 (0.15)	0.20 (0.13)	0.20 (0.13)	0.0	-	-	-	-
HF_LOW	1	0.90 (0.09)	0.90 (0.09)	0.70 (0.14)	0.40 (0.15)	0.10 (0.09)	0.10 (0.09)	0.10 (0.09)	0.10 (0.09)	0.0	-	-	-	-	-	-
E&P_HIGH	1	1	0.80 (0.13)	0.70 (0.14)	0.60 (0.15)	0.60 (0.15)	0.40 (0.15)	0.30 (0.14)	0.10 (0.09)	0.0	-	-	-	-	-	-
E&P_LOW	1	1	1	0.90 (0.09)	0.80 (0.13)	0.80 (0.13)	0.80 (0.13)	0.60 (0.15)	0.50 (0.16)	0.50 (0.16)	0.50 (0.16)	0.50 (0.16)	0.40 (0.15)	0.30 (0.14)	0.20 (0.13)	0.0

* Kaplan Meier and Mantel-Cox post-hoc test;

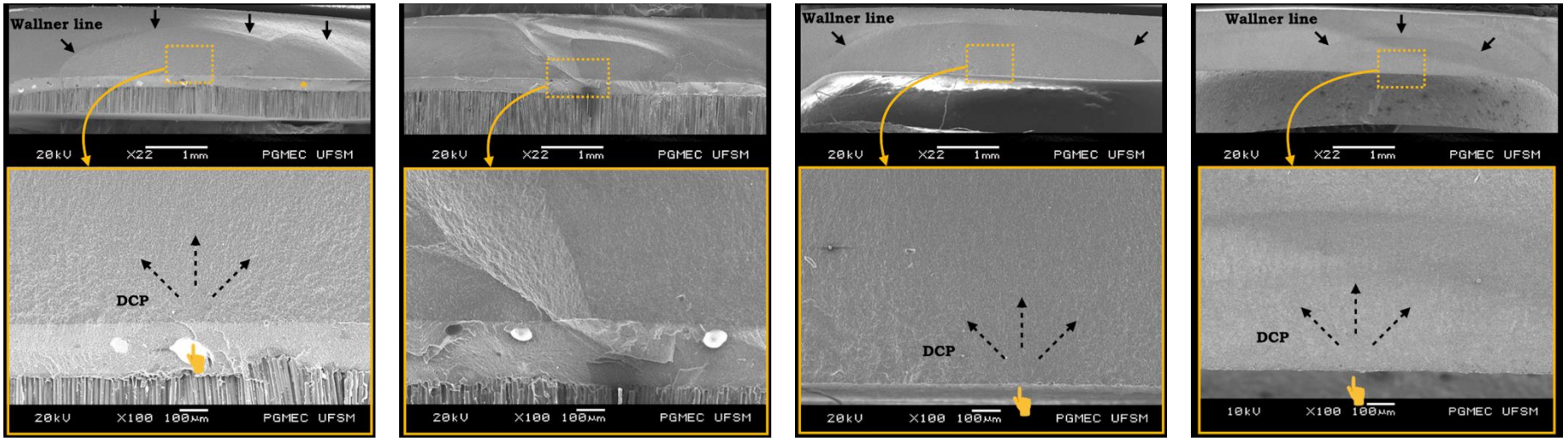
** The '-' symbol indicates that all specimens at the level (load/cycles) failed.

FIGURES



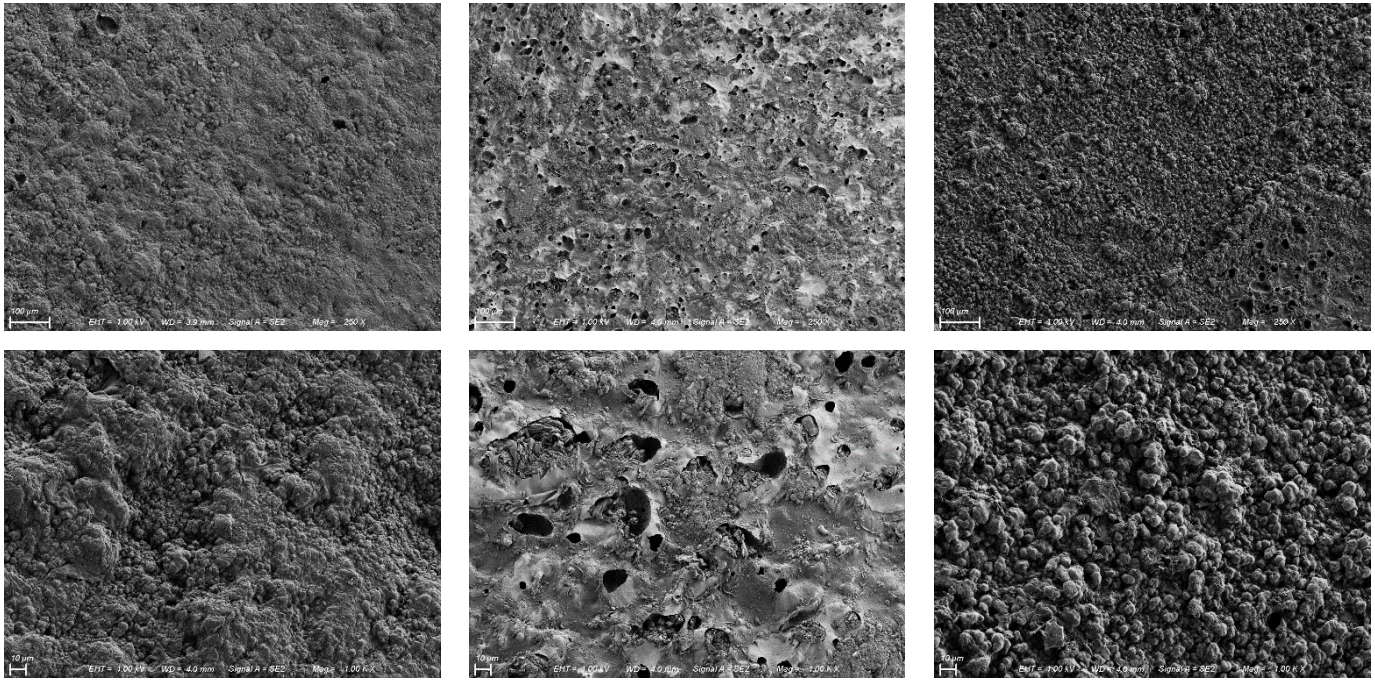
HF_HIGH HF_LOW E&P_HIGH E&P_LOW

Figure 1. Images representing transillumination of the failures found in the fatigue test. The yellow pointer indicates the radial cracks of each experimental group.



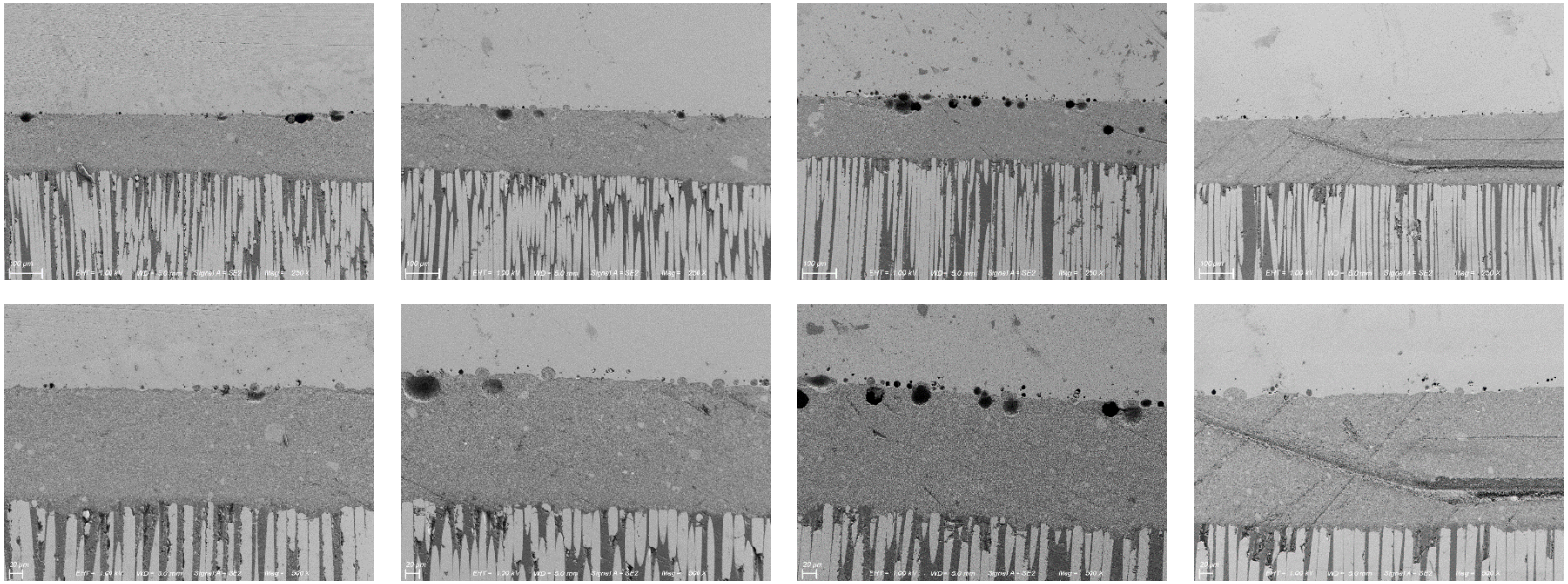
HF_HIGH HF_LOW E&P_HIGH E&P_LOW

Figure 2. SEM micrographs (22× and 100× magnification) illustrating the fractographic pattern accessed at one representative specimen of each experimental group. The yellow pointer indicates the origin of failure, while the dashed black arrows indicate the direction of the crack propagation (DCP); and the filled arrows indicate the Wallner lines (when present). At the HF_LOW group, a secondary crack was observed, pointing to more than one origin of failure.



As-machined HF E&P

Figure 3. Representative FE-SEM micrographs (250× and 1,000× magnification) of surface treatments: as-machined (non-etched), in order to compare the introduced defects by etching strategies, hydrofluoric acid + silane coupling agent (HF), and self-etching ceramic primer (E&P). Etched surfaces (HF and E&P) show a pattern alteration generated by the machined procedure. HF acid presents a more porous and irregular surface with larger and deeper voids compared to E&P, which is more homogenous and has a shallower pattern.



HF_HIGH HF_LOW E&P_HIGH E&P_LOW

Figure 4. Representative FE-SEM micrographs (250× and 500× magnification) illustrate the interface among the substrate-resin cement-ceramic and revealed unfilled areas at the cementation interface. There are irregularities noted in the intaglio surface of the etched ceramic where the resin cements were not able to properly fill or present bubbles, specifically in the HF_LOW and E&P HIGH.

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6 ARTIGO 5: Ceramic surface conditioning, resin cement viscosity, and aging relationships affect the load-bearing capacity under fatigue of bonded glass-ceramics

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Ceramic surface conditioning, resin cement viscosity, and aging relationships affect the load-bearing capacity under fatigue of bonded glass-ceramics

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Running title: Fatigue behavior of bonded glass-ceramics

Ceramic surface conditioning, resin cement viscosity, and aging relationships affect the load-bearing capacity under fatigue of bonded glass-ceramics

Abstract

This study aimed to evaluate the influence of ceramic surface treatments, resin cement viscosities, and storage regimens on the fatigue performance of bonded glass-ceramics (lithium disilicate, LD; feldspathic, FEL). Ceramic discs (Ø= 10 mm; thickness= 1.5 mm) were allocated into eight groups per ceramic (n= 15), considering three factors: "ceramic surface treatment" in two levels – 5% hydrofluoric acid etching and silane-based coupling agent application (HF), or self-etching ceramic primer (E&P); "resin cement viscosity" in two levels – in high or low viscosity; and "storage regimen" in two levels – baseline, 24 h to 5 days; or aging, 180 days + 25,000 thermal cycles. Adhesive luting was performed onto glass fiber-reinforced epoxy resin discs (Ø= 10 mm; thickness= 2 mm) and the bonded assemblies were subjected to cyclic fatigue tests: initial load= 200 N; step-size= 25 N (FEL) and 50 N (LD); 10,000 cycles/step; 20 Hz. Scanning electron microscopy (SEM) inspections were performed. Regarding the LD ceramic, the fatigue behavior was reduced after aging for HF_HIGH and E&P_LOW conditions, while stable performance was observed for HF_LOW and E&P_HIGH. Regarding the FEL results, aging negatively affected HF_HIGH, E&P_HIGH, and E&P_LOW, being that only the HF_LOW condition presented a stable behavior. The failure initiated from defects on the etched surface of the ceramics, where the cross-sectional analysis commonly revealed unfilled areas. Long-term aging might induce a decrease in mechanical behavior. The 'ceramic microstructure/surface conditioning/resin cement viscosity relationships' modulate the fatigue performance of lithium disilicate and feldspathic glass-ceramics

Keywords: Cementation. Dental bonding. Feldspathic. Lithium disilicate. Surface conditioning. Survival. Thermocycling.

Highlights

- The mechanical behavior of glass-ceramics bonded sets is influenced by their microstructure;
- Aging can negatively impact the mechanical performance of glass-ceramic bonded sets.
- High viscosity showed improved fatigue behavior with the ceramic primer on lithium disilicate;
- Both surface treatments can reach similar fatigue behavior with low viscosity for lithium disilicate;
- Both surface treatments and resin cement viscosities can be used to feldspathic after aging.

1. Introduction

Advances in computer-aided design and manufacturing (CAD/CAM) technology have boosted the development of all-ceramic dental restorations with superior biomechanical properties (Griggs, 2007; Spitznagel et al., 2018). Intense research efforts are underway to improve the resistance, aesthetics, and ability to reliably bond to dental substrates over time (Kelly and Benetti, 2011; Tian et al., 2014; Venturini et al., 2019). The most esthetic dental ceramics present an amorphous microstructure and contain high volumes of glassy phases (silica-based ceramics), such as feldspathic (40% volume of fillers), and can be reinforced by embedding crystals in the composition, such as lithium disilicate (70% volume of crystals), (Kelly and Benetti, 2011).

Based on a systematic review of the literature, monolithic ceramic restorations are considered an encouraging treatment for toothsupported single crowns and fixed partial dentures. Even with low failure rates (1–5%), minor biological and technical complications are reported, including restoration debonding (retention loss) (Mazza et al., 2022). In fact, the mechanical performance of ceramic crowns is impaired when the adhesion is negatively affected (da Rosa et al., 2022). Besides that, compromising the adhesion between tooth analogue substrate and ceramic induces a significant reduction in the fatigue behavior of lithium disilicate crowns, strengthening the importance of long-lasting adhesion for the longevity of the restorations (Chiapinotto et al., 2022).

Treatments of the bonding (intaglio) surface of the ceramic restorations have been proposed for adhesion improvements in glass-ceramics because surface physical alterations (a micromechanical interlocking mechanism) and chemical activation (bonding promoters) for adhesion to ceramic and resin materials through siloxane bonds are mandatory for stable mechanical performance (Manso et al., 2011; Tian et al., 2014; Sato et al., 2016; Venturini et al., 2015; Prochnow et al., 2018b; Scherer et al., 2018; Dapieve et al., 2020). These dual bonding mechanisms can be achieved by 4–10% hydrofluoric acid (HF) etching during 20 s–2 min depending on the type of glass-ceramic, followed by the application of a coupling agent containing silane (Tian et al., 2014; Dimitriadi et al., 2019). The HF introduces the surface changes necessary for resin cement infiltration and the silane enhances chemical interactions between the substrates (Tian et al., 2014; Dimitriadi et al., 2019). In addition to this well-established protocol, the etching/priming of the ceramic surface can be performed by applying a self-etching ceramic primer (E&P), (El-Damanhoury and Gaintantzopoulou, 2018; Prado et al., 2018; Scherer et al., 2018; Schestatsky et al., 2019; Dapieve et al., 2020, 2022). The E&P technique presents advantages such as simplification of the technique (single step), lower toxicity compared to HF, and satisfactory results (absence of cracks or debonding) in short-term clinical reports (Souza et al., 2020; Nascimento et al., 2021).

Milling and surface treatments can introduce microcracks in critical loaded areas and weaken glass-ceramics (May et al., 2021; Pilecco et al., 2021). Considering that ceramics have extremely low damage tolerance to failure (Kelly et al., 2017), the luting agent (resin cement) should be able to infiltrate and seal surface defects, reducing the flaw population and strengthening the ceramic structure (Naves et

al., 2010; Spazzin et al., 2016; Spazzin et al., 2017; Monteiro et al., 2018; Venturini et al., 2018; Dapieve et al., 2020). In this context, highly filled resin-based luting agents present higher viscosity and may differ from less viscous materials with regard to their potential for filling potential defects and distinct surface etching patterns (HF or E&P) (Spazzin et al., 2017; Scherer et al., 2018; Schestatsky et al., 2019; Dapieve et al., 2022).

Furthermore, the introduction and presence of surface flaws play a dominant role in damage accumulation, reduced lifetime, and fatigue behavior of ceramic structures (Kelly et al., 2017; Valandro et al., 2023). Indeed, restorations subjected to a complex environment in the oral cavity are influenced by several extrinsic factors that can stimulate the degradation (aging) of the materials involved in the restorative assembly (Lu et al., 2013; Tian et al., 2014). Aging procedures can lead to damage accumulation, which is important to understand the process of strength degradation (Kelly et al., 2017). Furthermore, according to Venturini and collaborators (2019), the fatigue performance of CAD/CAM ceramic restorative materials seems to be strongly influenced by the crystalline content immerged in the glassy matrix, highlighting the relevance of aging and fatigue tests in this context.

Research to date has not yet evaluated the effect of ceramic surface treatments and resin cement viscosity after aging effects on the fatigue behavior of glass-ceramics. Based on this, the aim of this study was to investigate the influence of ceramic surface treatments (HF acid etching + silane application Vs self-etching ceramic primer), resin cement viscosities (high vs. low), and storage regimens (baseline vs. aging) on the load-bearing capacity under fatigue of adhesively bonded glass-ceramics (lithium disilicate or feldspathic). The null hypotheses assumed were that (1) surface treatments, (2) resin cement viscosities, and (3) storage regimes would not affect the fatigue behavior of both bonded ceramics.

2. Material and methods

The general description of the materials used in the present study is listed in Table 1.

2.1 Study design

This study was designed in three factors for each lithium disilicate and feldspathic glass-ceramics (eight testing groups for each ceramic, n= 15, Table 2):

(i) "**Ceramic surface treatment**" in two levels: 5% hydrofluoric acid etching followed by a silane-based coupling agent application – HF groups; or self-etching ceramic primer – E&P groups;
(ii) "**Resin cement viscosity**" in two levels: high or low viscosity;
(iii) "**Storage regimen**" in two levels: baseline – 24 h up to 5 days of storage (distilled water, 37 °C); or aging – 180 days of storage (distilled water, 37 °C) plus 25,000 thermal cycles.

Bonded assemblies were used as a sample unit (Chen et al., 2014; Velho et al., 2022). They consisted of ceramic discs (Ø= 10 mm; thickness= 1.5 mm), which were adhesively luted onto substrate discs (thickness= 2.0 mm; Ø= 10 mm), reaching a final thickness of 3.5 mm. A simulation of the

topography pattern promoted by computer-aided design/computer-aided manufacturing (CAD/CAM) of the CEREC system was performed on the intaglio surface of ceramic discs (Rodrigues et al., 2018; Dapieve et al., 2020; Pilecco et al., 2021).

A schematic drawing of specimen preparation and fatigue testing setup is shown in Fig. 1.

2.2 Specimen preparation

As substrate (foundation), glass fiber-reinforced epoxy resin discs (thickness= 2.0 mm; Ø= 10 mm) were produced from plates (150 × 350 × 2.0 mm; Carbotec GmbH & Co. KG, Königs Wusterhausen, Germany), which were shaped into cylinders using a diamond drill (internal diameter= 10 mm; Diamant Boart, Brussels, Belgium) coupled to a bench drill (SBE 1010 Plus, Metabo, Nürtingen, Germany) with under water-cooling. The discs were manually polished on both sides with grit silicon carbide papers (SiC, #400- and #1200-grit) to remove irregularities, and then cleaned in an ultrasonic bath (distilled water; 5 min).

As ceramic restorations, lithium disilicate (IPS e.max CAD, Ivoclar, Schaan, Liechtenstein) and feldspathic CAD/CAM blocks (VitaBlocks Mark II for CEREC/inLab, Vita Zahnfabrik, Bad Säckingen, Germany) were manually shaped into cylinders (Ø = 10 mm) using a polishing machine with under water- cooling (EcoMet/AutoMet 250, Buehler, Lake Bluff, USA) with a sequence of silicon carbide papers (SiC, #80-, #400-, #600-, #1200-grit). Then, the cylinders were cut (Isomet 1000, Buehler) with under water-cooling, resulting in 240 ceramic discs (120 – lithium disilicate; 120 – feldspathic). They were polished with light digital pressure in a polishing machine (EcoMet/AutoMet 250, Buehler) on both surfaces with #120-, #400, and #1200-grit SiC papers until reaching a thickness of 1.5 mm and standardized surfaces.

After polishing, the bonding surfaces of the ceramic discs were subjected to an in-lab simulation of the CAD/CAM milling roughness of the CEREC system following previous literature (Rodrigues et al., 2018; Dapieve et al., 2020; Pilecco et al., 2021). A standardized size (100 mm × 50 mm) of #60 grit humidified SiC paper was used for each disc applying light digital pressure for 15 s on each axis (x and y). After that, the feldspathic remained untouched and lithium disilicate discs were crystallized (Vacumat 6000 MP, VITA Zahnfabrik) as recommended by the manufacturer (840 °C, 7 min vacuum). Next, the roughness values of all samples were measured to compare them to those generated by CAD/CAM machining (Carrabba et al., 2017; Fraga et al., 2017), as well as to certify that all groups received similar roughness prior to the surface treatments investigated herein. For this purpose, six measurements were performed for each specimen (axes x and y) on a contact profilometer (Mitutoyo SJ-410, Mitutoyo Corporation, Kawasaki, Japan). The roughness (mean and standard deviation) achieved by the in-lab simulation for Ra and Rz (μm) parameters were: Ra: 1.76 μm, SD: 0.15; Rz: 11.20, SD: 0.85 for lithium disilicate, being compatible with the one generated by CAD/CAM machining (Ra: 1.84 μm, SD: 0.18; Rz: 11.07 μm, SD: 1.00) obtained by Fraga et al. (2017) and Ra: 1.09 μm, SD: 0.11 for feldspathic, being compatible to the one generated by CAD/CAM machining (Ra: 1.08 μm, SD

0.18) obtained by Carrabba et al. (2017). The specimens were subsequently cleaned (isopropyl alcohol, 5 min) in an ultrasonic bath (1440 D, Odontobras, Ribeirão Preto, Brazil).

2.3 Ceramic surface treatments

The bonding surfaces of the ceramic discs received one of the following surface treatments according to the study design:

(i) **HF groups**: 5% hydrofluoric acid (IPS Ceramic Etching Gel, Ivoclar) was applied with a microbrush for 20 s (lithium disilicate) and 60 s (feldspathic), removed with air-water spray for 30 s, and air-dried for 30 s. Next, the etched surface received a silane-based coupling agent (Monobond N, Ivoclar), which was actively scrubbed for 15 s and allowed to react for another 45 s, with posterior air-dry for 30 s.

(ii) **E&P groups**: a self-etching ceramic primer (E&P, Monobond Etch & Prime, Ivoclar) was rubbed on the ceramic surface using a microbrush for 20 s actively, left to react for 40 s, air-water sprayed for 30 s, and air-dried for 30 s, featuring a simplified conditioning system (Scientific Documentation; Scherer et al., 2018; Dapieve et al., 2020; Dapieve et al., 2022).

All discs were subjected to an ultrasonic bath (distilled water up to 5 min) after the ceramic surface treatments (i.e., after HF and one-step E&P).

2.4 Bonding procedures

The bonding surfaces of glass fiber-reinforced epoxy resin discs were etched with 10% hydrofluoric acid (Condac Porcelana, FGM, Joinville, Brazil) for 1 min, followed by rinsing with air-water spray (30 s), air spray (30 s), ultrasonic cleaning (5 min, distilled water) and adhesive (ExciTE F DSC, Ivoclar, Variolink system) application for 10 s, according to the cement manufacturer's recommendation. Next, a dual-curing resin cement (Variolink N, Ivoclar) in high or low viscosity (catalyst) was mixed at a 1:1 ratio with the cement base and applied to the treated surfaces of the ceramic discs. Each ceramic disc was adhesively luted to a substrate disc under a constant load of 2.5 N for 10 min. The resin cement excesses were subsequently removed and the assemblies were light-cured (Radii-cal LED curing light, SDI, Bayswater, Australia) for five exposures of 20 s each (one in each direction of 5 positions: 0°, 90°, 180°, 270°, and on top).

2.5 Storage regimens

The bonded assemblies of each condition were randomly assigned to the two storage regimen conditions (Table 2):

(i) **Baseline**: storage in distilled water at 37°C for approximately 24 h until 5 days (variance based on the time necessary to execute the fatigue test);

(ii) **Aging**: storage in distilled water at 37°C for 180 days and 25,000 thermal cycles (Nova Ética, São Paulo, Brazil) of 30 s baths at 5 and 55 °C and transfer time of 5 s (Armstrong et al., 2017; Van Meerbeek and Frankenberger, 2020).

2.6 Cyclic fatigue test

The bonded assemblies (n= 15) were tested using a cyclic fatigue test (Kelly et al., 2017, Velho et al., 2022) in an electric machine (Instron ElectroPuls E3000, Instron, Norwood, United States). Cyclic loads were applied with a 40 mm diameter stainless-steel hemispheric piston (Kelly et al., 2010) under distilled water at a frequency of 20 Hz (Velho et al., 2020). An adhesive tape (110 µm) was placed between the piston and the ceramic surface to reduce contact stress concentration (Kelly, 1999). An initial load of 200 N for 10,000 cycles was performed and incremental steps of 50 N (lithium disilicate assemblies) or 25 N (feldspathic assemblies) for 10,000 cycles were applied until failure (radial cracks) of the sample. The specimens were checked for cracks at the end of each step by light oblique transillumination and the corresponding failure data (load and number of cycles to failure) of each sample were recorded for statistical analysis.

2.7 Fractographic analysis

All the specimens were inspected by stereomicroscope (Discovery V20, Carl Zeiss, Gottingen, Germany) after the fatigue test, and representative samples (n= 1) were selected from each ceramic material, in which the ceramic fragments were detached to access the origin of the defects. The fragments were ultrasonically cleaned (78% isopropyl alcohol, 5 min), air-dried, gold-sputtered, and analyzed under scanning electron microscopy (SEM, Vega3, Tescan, Brno, Czech Republic) at 200× and 1,000× magnifications to determine the crack origin characteristics.

2.8 Topographic analysis

Additional samples of each ceramic were produced to be inspected regarding the topographical changes, microstructure features, and alterations after the surface treatments. The surfaces of the specimens were treated according to the above described, gold-sputtered, and analyzed by SEM (Vega3, Tescan) at 100× and 5,000× magnifications.

2.9 Cross-sectional bonded interfacial zone analyses

One specimen per group was randomly selected and transversely sectioned in a cutting machine (Isomet 1000, Buehler) to inspect the morphology of the bonding interfaces, the defects introduced by the treatments, and the filling of these defects by the different viscosities of the resin cement. Then, the cross-section surface was mirror-polished (EcoMet/AutoMet 250, Buehler) using #400-, #600-, #1200- and #2200 grit silicon carbide papers, cleaned in an ultrasonic bath (78% isopropyl alcohol, 5 min), air-dried, gold-sputtered, and analyzed by SEM (Vega3, Tescan) at 5,000 × magnification.

2.10 Data analyses

A statistical software program (IBM SPSS Software, IBM, Armonk, United States) was used to perform the analysis with a significance level of 0.05. Data were diagnosed as non-normal by the Shapiro-Wilk test (p= 0.002 for both ceramics). Fatigue data were subjected to non-parametric survival analysis (Kaplan Meier and Mantel-cox post-hoc tests) to access the mean and the confidence intervals of fatigue failure load (FFL), cycles for failure (CFF), and survival rates. Due to non-normal data distribution, a factorial analysis of variance (ANOVA 3-way) with bootstrapping correction (1000 re-samplings; 95% CI, bias-corrected and accelerated, BCa) was performed to determine the influence of each factor (ceramic surface treatment, resin cement, and storage regimen) and the interactions among them. SEM analyses were qualitatively/descriptively evaluated.

3. Results

Fatigue data (Table 3, Fig. 2) demonstrated that lithium disilicate etched with hydrofluoric acid and low viscosity resin cement showed the lowest FFL and CFF values (LD/HF_LOW) at the baseline condition; however, the mechanical performance was similar to LD/HF_HIGH. The deleterious impact of aging can be observed in the LD/HF_HIGH and LD/E&P_LOW groups; thus, the lowest values of FFL and CFF were detected in the LD/HF_HIGH group, although presenting statistical similarities to HF_LOW and E&P_LOW. With the use of high viscosity cement, conditioning with a self-etching ceramic primer presented the best mechanical results, while bonding with low viscosity cement, the behavior was similar between the two surface treatments (HF and E&P).

For feldspathic ceramics, high viscosity resin cement with the tested surface treatments (HF and E&P) performed better than low viscosity (HIGH viscosity > LOW viscosity; HF = E&P) at baseline conditions and the aging negatively affected the performance of FEL/HF_HIGH, FEL/E&P_HIGH, and FEL/E&P_LOW. After aging procedures, all interactions (HF/E&P surface treatments; high/low viscosities) led to similar results among groups.

Three-way ANOVA (Table 4) showed a statistically significant influence ($p < 0.05$) of surface treatment, aging, and the interaction among storage regimen*resin cement*surface treatment for FFL and CFF outcomes regarding lithium disilicate ceramic. For feldspathic ceramic, storage regimen, resin cement, and surface treatment presented statistical significance when evaluated separately; however, the interaction among those three study factors is not statistically significant (p= 0.346 for FFL; p= 0.376 for CFF).

The fractographic analysis showed that the failures (all of them - radial cracks) originated from the defects located on the tensile side of the lithium disilicate and feldspathic ceramics (i.e., on the bonding surface) (Fig. 3). The micrographs demonstrated that HF and E&P surface treatments promoted topographic alterations at different intensities. A large number of defects were observed after HF acid etching for both ceramics, and specific topographic patterns were identified for the different glass-

ceramics. HF promoted an irregular topographic pattern with deeper defects compared to E&P, which had a shallower pattern (Fig. 4).

SEM of the cross-sectional bonded area revealed filled interfaces for lithium disilicate ceramic and unfilled zones between feldspathic ceramic and the resin cements. It is also possible to observe some differences regarding the resin cement thickness (Fig. 5).

4. Discussion

Overall, the present study strengthens the idea that a host of variables, including ceramic microstructure, surface treatments, different viscosities of luting agents, and storage regimens over time can influence the mechanical performance of bonded glass-ceramics. Regarding bonded lithium disilicate ceramic under aging conditions, a high viscosity resin cement presented the best fatigue performance with the self-etching ceramic primer treatment, while a low viscosity resin cement presented similar behavior when considering the two surface treatments evaluated (HF followed by silane and self-etching ceramic primer). Regarding bonded feldspathic ceramic after aging, both surface treatments and resin cement viscosities can be used to reach a similar mechanical behavior. Therefore, the hypotheses that surface treatments, resin cement viscosities, and storage regimes would similarly affect fatigue behavior were partially accepted.

Firstly, the lithium disilicate and the feldspathic ceramics showed distinct behaviors for the bonding procedures variables; that is, they did not show a similar trend in mechanical behavior (Table 3, Fig. 2). These findings mainly reflect the specific microstructure of the ceramics (Zhang and Kelly, 2017; Hallmann et al., 2018; Venturini et al., 2019). In this sense, several factors can affect the ceramic bonded sets evaluated, including different surface states (Zhang and Kelly, 2017), topography changes after each surface treatment (Fig. 4; Zhang and Kelly, 2017; El-Damanhoury and Gaintantzopoulou, 2018; Scherer et al., 2018; Schestatsky et al., 2018; Dapieve et al., 2021; Dapieve et al., 2022), distinct resin cement abilities to penetrate and fill the surface defects (Fig. 5; Barbon et al., 2019; Dapieve et al., 2022), and storage regimens (Lu et al., 2013; Scherer et al., 2018; Dapieve et al., 2020).

Differences in terms of microstructure can influence flaw distribution, elastic properties, and other mechanical properties of glass-ceramics (Griggs, 2007; Trindade et al., 2016; May et al., 2021). Indeed, the microstructure is the prominent factor that affects the mechanical properties of lithium disilicate (Hallmann et al., 2018). Etching by 5% HF induces a rod-like morphology with the interlocking microstructure of the lithium disilicate phase (Fig. 4), (Zhang and Kelly, 2017; El-Damanhoury and Gaintantzopoulou, 2018; Hallmann et al., 2018; Scherer et al., 2018; Dapieve et al., 2021; Dapieve et al., 2022), while E&P self-etching ceramic primer generates a shallower pattern compared to HF etching (El-Damanhoury and Gaintantzopoulou, 2018; Scherer et al., 2018; Schestatsky et al., 2018; Dapieve et al., 2021; Dapieve et al., 2022), which are in agreement with the factorial data on FFL and CFF (Table 4). Even given these surface characteristics of the lithium disilicate ceramic, after resin bonding to a substrate, the defects are filled by both luting agents, as seen in Fig. 5, leading to the understanding that

cements of different viscosities were able to penetrate the conditioned surfaces of the ceramic discs. In contrast, a study in the same line of research (Dapieve et al., 2022) found differences in filling defects using the same resin cements of different viscosities and surface treatments (HF and E&P). These differences can be explained by the surface topographies (CAD/CAM simulated and tested herein vs. machined, Dapieve et al., 2022) and sample geometry (disc tested herein vs. crown, Dapieve et al., 2022), which may have influenced the mechanical outcomes (Velho et al., 2022) and the bonding procedure during specimen seating.

In a non-aged condition (*baseline*), a high viscosity resin cement associated with both surface treatments presented a similar fatigue performance. Furthermore, a low viscosity cement promoted improved mechanical behavior with the self-etching ceramic primer agent, whose findings are in agreement with the previous study cited above (Dapieve et al., 2022). After aging protocols (180 days of water storage and 25,000 thermal cycles; Armstrong et al., 2017; Van Meerbeek and Frankenberger, 2020), a deleterious effect on fatigue performance can be observed in some groups (Table 3; Table 4; Fig. 2), which is a prominent outcome due to the importance of aging regimen and its effects on adhesion durability and its consequences on the threshold mechanical performance of the bonded assembly. Considering that aging in water does not affect the flexural strength of glass-ceramics (lithium disilicate - e.max CAD; and feldspathic - Vita Mark II; Kim et al., 2020; Sonmez et al., 2018), the storage regimen (aging) can influence the bonded interfacial zone (resin cement interface) (Lu et al., 2013). In this sense, considering the statistically significant interaction for storage regime*resin cement*surface treatment (Table 4), two groups of lithium disilicate bonded ceramics showed a degradation of the fatigue outcome after aging (LD/HF_HIGH and LD/E&P_LOW), with the worst values for the group treated with HF and luting with high viscosity resin cement. The former result may be related to larger and deeper voids caused by HF etching (Scherer et al., 2018), which were partially filled by a high viscosity resin cement. More filler particles in high viscosity cement (+8.4 vol.%; +6 wt.%) and less monomer content (-5.9 wt.%) can stimulate swelling processes, plasticization, and the possibility of resin structure decomposition (Niem et al., 2020), damaging the interaction between etching surface pattern and the resin cement after aging.

Also related to resin cement aging, a prior study by Lu et al., (2013) found that water aging (30 days – a shorter period compared to our study, which was 180 days + thermos cycles) promotes a reduction of stiffness and bonding strength of a resin cement agent. The authors suggested that degradation in bonding strength and stiffness could potentially lead to stress redistribution in the restored crown and reduce the load-bearing capacity of all-ceramic restorations, which can also explain the performance of aged groups in our study.

With regard to the bonded feldspathic ceramic, 5% HF etching resulted in honeycomb-like porous surfaces consisting of gross irregularities and pores, while self-etching generated a less pronounced etching pattern (and less rough) than HF (Fig. 4) (El-Damanhoury and Gaintantzopoulou, 2018) with a statistically significant effect of surface treatment factor for FFL and CFF (Table 4). According to the

cross-sectional bonded interfacial zone analyses (Fig. 5), unfilled areas at the cementation interface of feldspathic ceramic are noted. Our findings are in agreement with Venturini et al. (2018), who observed that defects promoted by low hydrofluoric acid concentrations (such as 1% and 5%) were not entirely filled by the resin cement, while the resin cement penetrated into the irregularities (apparently larger and rounded) introduced by 10% HF. Indeed, this difficulty of resin cement in filling surface defects may be associated with the differentiated microstructure of feldspathic ceramic. Therefore, higher stress concentration around the existing defects can lead to premature fracture during smaller loads (Venturini et al., 2018).

The baseline results of bonded feldspathic showed the best fatigue performance for the high viscosity resin cement ($p<0.05$; resin cement factor, Table 4) for both surface treatments. Indeed, those findings are in line with Barbon et al. (2019), who investigated experimental resin-based luting agents with distinct fillers loading, and demonstrated that resin cement with high inorganic filler content strengthened a bonded feldspathic ceramic etched with 10% hydrofluoric acid. In addition, an increment in filler content was positively associated with the flexural and characteristic strengths of the bonded specimens (Barbon et al., 2019). Hence, studies have shown that the elastic modulus of the luting agent is positively associated with the strengthening of glass-ceramics (Spazzin et al., 2016; Spazzin et al., 2017). However, after aging (a statistically significant factor, Table 4), it was not found that all groups with high viscosity presented the best results, as both surface treatments and resin cement viscosities resulted in a similar mechanical performance of the bonded feldspathic (Table 3; Fig. 2). This fact can be also explained by the resin cement composition - as mentioned above, fillers of higher viscosity cement and less monomer content can stimulate the degradation (Niem et al., 2020). Lastly, the results corroborate the idea that, in our context, the mechanical behavior depends on different variables: ceramic microstructure, ceramic surface conditioning method, resin agent viscosity, and, finally, storage conditions.

Another factor that can be related to the resin cement viscosity is the resin cement thickness (Barbon et al., 2019). Lithium disilicate crowns withstood higher loads when bonded with a 50 µm layer compared to the 500 µm one (May et al., 2012; Gressler May et al., 2015). Studies support that thinner cement thickness is significantly related to mechanical behavior improvement (Silva et al., 2008; Rojpaibool and Leevailoj, 2017). On the other hand, there is also the indication that small differences in layer thickness may not influence the stress distribution (60 – 120 µm, Tribst et al., 2021; 100 – 300 µm, Tribst et al., 2018), bond strength (60 – 120 µm, Tribst et al., 2021), fracture resistance - monotonic (50 – 100 µm, May et al., 2012; 50 – 100 µm, Rojpaibool and Leevailoj, 2017) or fatigue performance (50 – 300 µm, Venturini et al., 2020; 50 – 200 µm, Baldissara et al., 2021) of glass-ceramic assemblies. In fact, the cross-sectional micrographs showed some differences regarding the resin cement thickness by comparing the representative samples of each group (Fig. 5). It is not possible to verify if this difference in the cement layer influenced the mechanical performance in fatigue, and therefore, we consider it a limitation of the study, being a suggestion for future researches. Furthermore, by adding

fillers (increasing the viscosity), the elastic modulus (E) can be affected, even if the impact on the elastic modulus depends on a certain difference in the proportion of particles, i.e.: 55wt% resin cement presents a similar E to 65wt% resin cement and different compared to 75%wt one (Barbon et al., 2019).

Even with certain differences and with the care that an in vitro study should not be directly extrapolated to clinical practice, we can determine that the mean values of fatigue failure load regarding bonded lithium disilicate ceramic (890 – 977 N) are considered enough to withstand masticatory loads when in the oral cavity (mean total masticatory force of 220 N, with a maximum of 450 N; Morneburg and Pröschel, 2002). However, feldspathic restorations should be used with caution in the posterior region, taking into account the fatigue failure loads reached after aging (415 – 478 N).

The conditioning of the intaglio surface of glass-ceramic can be performed with HF followed by a coupling agent containing silane or self-etching ceramic primer. Considering the mechanical fatigue performance after aging, the resin bonding procedures can be performed with both surface treatments for feldspathic ceramics and with specific improved combinations for lithium disilicate. However, these findings should be evaluated with caution, as the study has features that may limit the data extrapolation. The major study limitation refers to the standardization of the thickness of the cement layer, once the same constant load of the bonding procedures for resin cement with different viscosities can affect the final film thickness and also the stress distribution. Besides that, the application of axial loads, the use of simplified disc samples (although this excludes confounding anatomy variables), and the use of analog material of dentin as a substrate need to be considered. Further investigation evaluating anatomic specimens, different resin cement compositions, and sliding load application are strongly recommended.

5. Conclusion

- The fatigue performance of lithium disilicate and feldspathic bonded sets are strongly influenced by their microstructure;
- Long-term aging (water storage and thermal cycles) might induce a decrease in the mechanical performance of glass-ceramic bonded sets.
- The high viscosity resin cement presented the best fatigue performance with the self-etching ceramic primer on bonded lithium disilicate assemblies. With a low viscosity resin cement, the interaction promotes similar mechanical performance between the two surface treatments evaluated (HF followed by silane-based coupling agent and self-etching ceramic primer).
- Considering bonded feldspathic after aging, both surface treatments and resin cement viscosities can be used to reach a similar mechanical behavior.

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Tables

Table 1. Description of materials, commercial name, manufacturer, composition, and batch number.

Material	Trade market and manufacturer	Main composition*	Batch number
5% HF	IPS Ceramic Etching Gel, Ivoclar, Schaan, Liechtenstein	< 5% Hydrofluoric acid	W14921
10% HF	Condac Porcelana, FGM, Joinville, Brazil	< 10% Hydrofluoric acid	220920
Dual-curing resin cement	Variolink N High Viscosity (Catalyst), Ivoclar	Barium glass filler and mixed oxide (52.2%**), dimethacrylates (22.0%), ytterbiumtrifluoride (25%), initiators and stabilizers (0.8%) and pigments (< 0.1%)	Y15347
	Variolink N Low Viscosity (Catalyst), Ivoclar	Barium glass filler and mixed oxide (46.2%), dimethacrylates (27.9%), ytterbiumtrifluoride (25%), initiators and stabilizers (0.9%) and pigments (< 0.1%)	Y06775
	Variolink N Base, Ivoclar	Barium glass filler and mixed oxide (48.4%), dimethacrylates (26.3%), ytterbiumtrifluoride (25%), initiators and stabilizers (0.3%) and pigments (< 0.1%)	Y06775
Dual-curing total-etch adhesive	ExciTE F DSC, Ivoclar	Phosphonic acid acrylate, dimethacrylates, hydroxyethyl methacrylate, highly dispersed silicon dioxide, ethanol, catalysts, stabilizers, and fluoride	X25002
Feldspathic ceramic	VITABLOCS Mark II, Vita Zahnfabrik, Bad Sackingen, Germany	Al_2O_3, SiO_2, K_2O, Na_2O, CaO, TiO_2, oxides	45950
Lithium disilicate ceramic	IPS e.max CAD, Ivoclar	SiO_2, Li_2O, K_2O, P_2O_5, ZrO_2, ZnO, Al_2O_3, MgO, oxides	X27104
Self-etching ceramic primer	Monobond Etch & Prime, Ivoclar	Ammonium polyfluoride, silane system based on trimethoxypropyl methacrylate, alcohols, water, and colorant	Y42802
Silane-based coupling agent	Monobond N, Ivoclar	Alcohol solution of silane methacrylate, phosphoric acid methacrylate, and sulfide methacrylate	Z00DTK

* The chemical composition is described according to the manufacturers' information;
** % in weight.

Table 2. Experimental design of the study (n= 15) considering two glass-ceramics, and the factors of the study: surface treatments, luting agents in different viscosities and storage conditions.

Glass-ceramic	Group	Surface treatment	Resin cement	Storage regimen
Lithium disilicate **LD**	**LD/HF_HIGH**	5% Hydrofluoric acid + silane-based coupling agent (HF)	High viscosity	Baseline*
				Aging**
	LD/HF_LOW		Low viscosity	Baseline
				Aging
	LD/E&P_HIGH	Self-etching ceramic primer (Monobond Etch and Prime, E&P)	High viscosity	Baseline
				Aging
	LD/E&P_LOW		Low viscosity	Baseline
				Aging
Feldspathic **FEL**	**FEL/HF_HIGH**	5% Hydrofluoric acid + silane-based coupling agent (HF)	High viscosity	Baseline
				Aging
	FEL/HF_LOW		Low viscosity	Baseline
				Aging
	FEL/E&P_HIGH	Self-etching ceramic primer (Monobond Etch and Prime, E&P)	High viscosity	Baseline
				Aging
	FEL/E&P_LOW		Low viscosity	Baseline
				Aging

***** 24 h up to 5 days of storage (distilled water, 37 °C);
****** 180 days of storage (distilled water, 37 °C) plus 25,000 thermal cycles.

Table 3. Mean of the fatigue test for fatigue failure load (FFL) in Newton and cycles for failure (CFF) with 95% confidence intervals depicted by Kaplan Meier and Mantel-Cox post-hoc test.

Group	Outcome	Lithium disilicate/LD		Feldspathic/FEL	
		Storage condition			
		Baseline	Aging	Baseline	Aging
HF_HIGH	FFL	990 (935 – 1,045)^{ABa}	890 (837 – 943)^{Bb}	562 (536 – 587)^{Aa}	478 (464 – 492)^{Ab}
	CFF*	168,000 (156,979-179,021)	148,000 (137,488 – 158,512)	154,667 (144,390 – 164,944)	121,333 (115,638 – 127,029)
HF_LOW	FFL	970 (923 – 1,017)^{Ba}	957 (920 – 993)^{ABa}	503 (478 – 528)^{Ba}	465 (427 – 503)^{Aa}
	CFF*	164,000 (154,669 – 173,331)	161,333 (153,958 – 168,708)	131,333 (121,236 – 141,431)	114,667 (101,572 – 127,761)
E&P_HIGH	FFL	1,023 (980 – 1,067)^{Aba}	977 (930 – 1,023)^{Aa}	550 (528 – 572)^{Aa}	457 (438 – 475)^{Ab}
	CFF*	174,667 (165,929 – 183,404)	165,333 (155,989 – 174,678)	150,000 (141,235 – 158,765)	112,667 (105,144 – 120,189)
E&P_LOW	FFL	1,050 (991 – 1,109)^{Aa}	910 (851 – 969)^{ABb}	497 (470 – 523)^{Ba}	415 (398 – 431)^{Ab}
	CFF*	181,333 (168,665 – 194,002)	152,000 (140,178 – 163,822)	128,667 (118,040 – 139,294)	96,000 (89,429 – 102,571)

* Note that the statistical significance for FFL and CFF is the same.

** Same uppercase letters on each column indicate statistical similarity among surface treatments and resin cements.

*** Same lowercase letters on each row indicate statistical similarity in the same group considering storage conditions (baseline and aging).

Table 4. Three-way ANOVA results of fatigue failure load (FFL) and cycles for failure (CFF) considering the three factors of the study and the interactions among them.

	Lithium disilicate				Feldspathic			
	FFL		CFF		FFL		CFF	
	F	p	F	p	F	p	F	p
Aging	16.951	0.000	17.258	0.000	70.904	0.000	78.914	0.000
Resin cement	0.008	0.927	0.033	0.857	22.379	0.000	25.340	0.000
Surface treatment	4.438	0.038	4.698	0.032	6.526	0.012	6.586	0.012
Aging*cement	0.008	0.927	0.033	0.857	2.587	0.111	2.494	0.117
Aging*treatment	1.013	0.316	1.174	0.281	2.292	0.133	2.192	0.142
Cement*treatment	1.415	0.237	1.174	0.281	0.439	0.509	0.351	0.555
Aging*cement*treatment	6.102	0.015	6.394	0.013	0.895	0.346	0.789	0.376

Figures

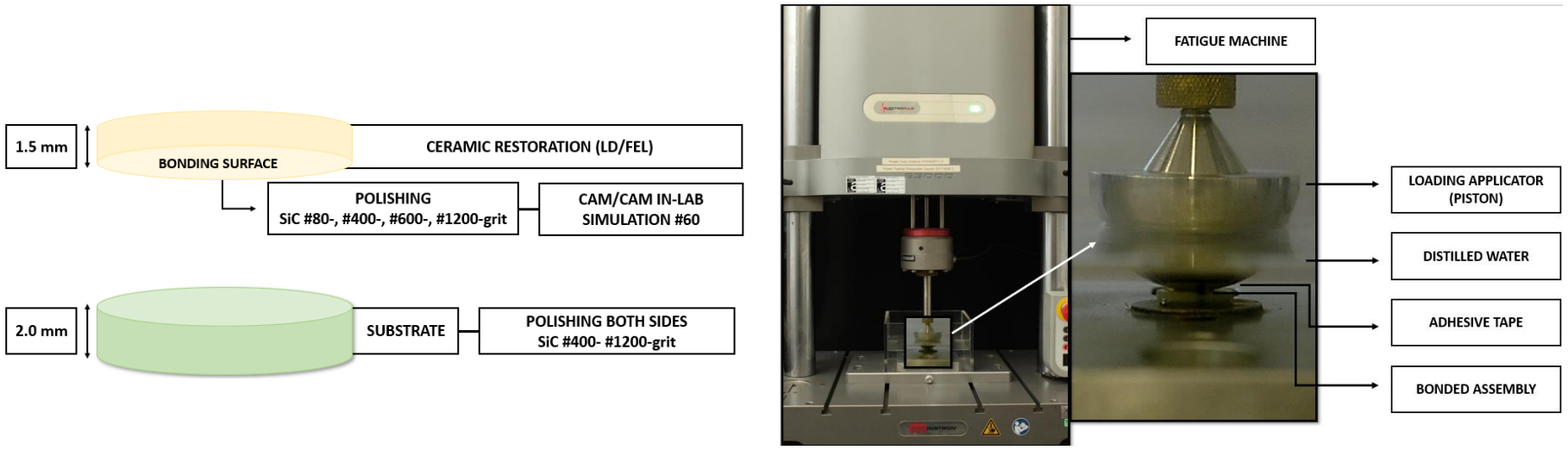
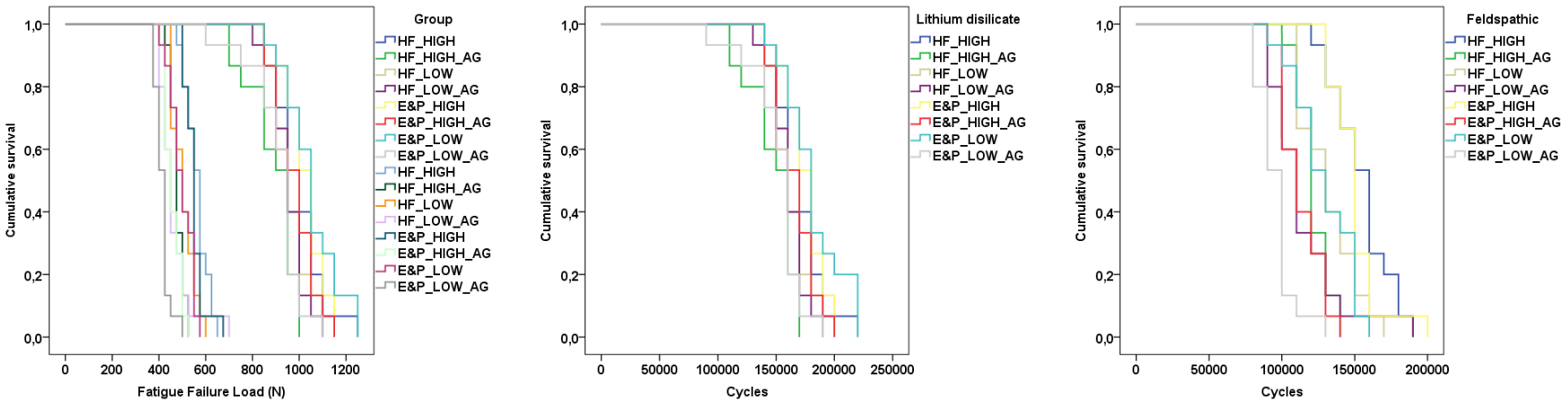


Figure 1. A) Specimen preparation of the simplified ceramic restorations and substrates; B) Cyclic fatigue test setup.



A **B** **C**

Figure 2. Survival graphs were obtained by the Kaplan-Meier and Mantel-Cox (log-rank) test. A) Fatigue failure load. On the left: feldspathic; on the right: lithium disilicate; B) Cycles for failure regarding lithium disilicate; C) cycles of feldspathic data. It should be noted that the graphs for cycles are not on the same scale (different load steps), showing that lithium disilicate has a high survival probability compared to feldspathic.

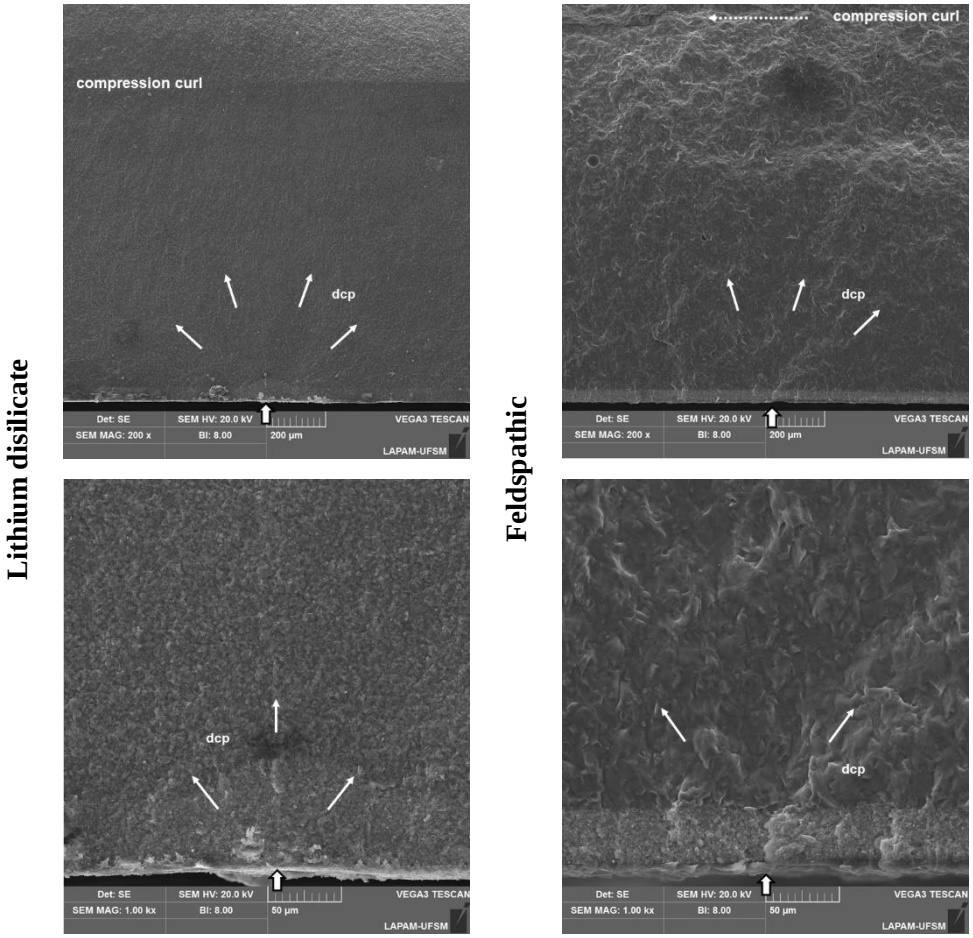
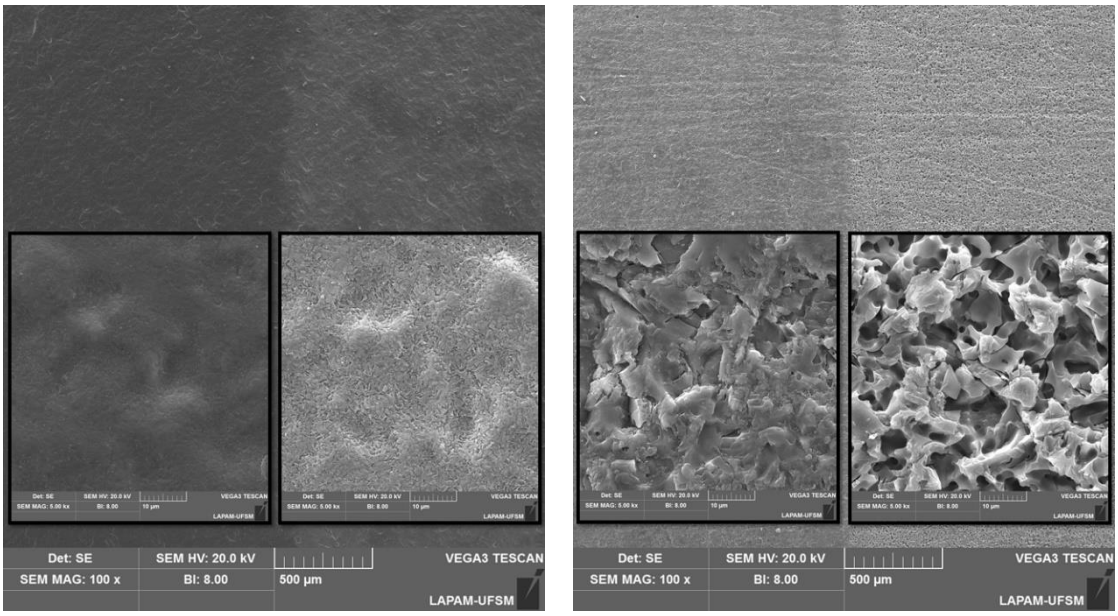


Figure 3. SEM micrographs (200× and 1000× magnification) illustrate the fractographic pattern accessed in a representative specimen of each ceramic material. The thick arrow in the bottom region indicates the origin of failure at the cementation interface from defects on the ceramic surface. Thin arrows indicate the direction of the crack propagation (dcp) towards the top surface, where the compression curl can be observed.



Lithium disilicate **Feldspathic**

Figure 4. SEM images (100× and 5,000× magnification) of the surface treatments were evaluated. The sample of each ceramic material was divided to observe distinct topographic changes: on the left of each micrograph, it is possible to observe the etched surface with self-etching ceramic primer (E&P, 60 s), and on the right with 5% hydrofluoric acid (HF, 20 s). HF acid suggests a more porous and irregular surface with larger craters and pits compared to E&P, which is slighter, in both ceramics. It is also possible to identify that the topographical pattern after HF etching is different between the two ceramics: elongated crystals (rod-like morphology) dispersed with shallow irregularities for lithium disilicate and honeycomb for feldspathic.

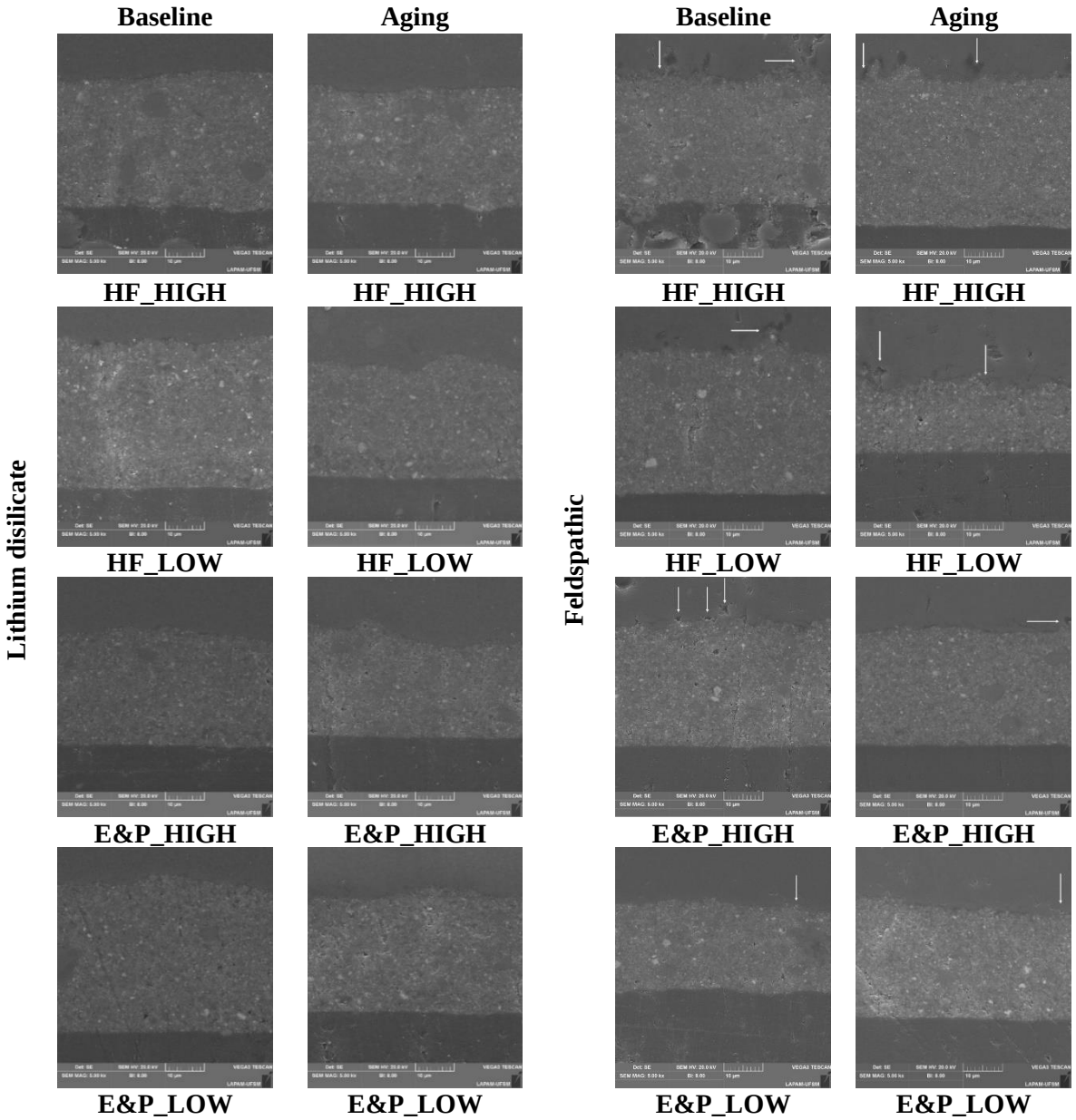


Figure 5. Representative SEM micrographs (5,000× magnification) demonstrate the ceramic (top) - resin cement (middle) - glass fiber-reinforced epoxy resin substrate (bottom) interface. In general, visual inspection allows to observe surface defects filled by both resin cements for lithium disilicate ceramics, while for feldspathic ceramics some unfilled defects (white arrows) can be detected at the interface between the ceramic and the resin cements. Besides that, it is possible to detect some resin cement thickness differences by comparing the specimens.

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7 CONSIDERAÇÕES FINAIS

Com base nas investigações da presente tese, evidencia-se o intertravamento micromecânico entre a superfície de cimentação da cerâmica e o agente cimentante resinoso como preponderante para o comportamento mecânico em fadiga de restaurações simplificadas de dissilicato de lítio. Observa-se ainda que, no cenário testado, o agente de união à base de silano aplicado após o condicionamento com ácido fluorídrico apresentou-se como um passo laboratorial dispensável.

Além disso, compreende-se que a alteração topográfica da superfície de cimentação de uma cerâmica vítrea associada a diferentes viscosidades de um agente cimentante resinoso podem apresentar desfechos distintos no que diz respeito à resistência de união ao dissilicato de lítio, de acordo com cada tratamento de superfície da cerâmica, regime de armazenamento e modo de aplicação de carga. Ressalta-se que a aplicação de cargas intermitentes (cisalhamento sob fadiga cíclica) pode impactar deleteriamente no comportamento adesivo e nas probabilidades de sobrevivência de conjuntos de dissilicato de lítio adesivamente cimentados, independentemente da viscosidade do cimento resinoso empregado.

Considerando um contexto mais complexo, o desempenho mecânico em fadiga de coroas usinadas de dissilicato de lítio adesivamente cimentadas depende substancialmente da topografia existente, dos tipos de defeitos e irregularidades da superfície interna e de como o agente de cimentação preenche as irregularidades introduzidas pelos tratamentos de superfície.

Em última análise, ratifica-se que o envelhecimento pode afetar negativamente o comportamento em fadiga de diferentes cerâmicas vítreas adesivamente cimentadas. Nesse sentido, as relações da microestrutura cerâmica, do tratamento de superfície e da viscosidade do agente cimentante parecem modular o desempenho em fadiga dos materiais testados.

Em suma, destaca-se que a alteração topográfica resultante do processamento e dos tratamentos de superfície prévios à cimentação (ácido fluorídrico seguido da aplicação de agente de união à base de silano ou primer cerâmico autocondicionante), a viscosidade do agente cimentante resinoso (alta ou baixa), o regime de armazenamento (com ou sem protocolos de envelhecimento), o modo de aplicação de carga (estática ou cíclica) e, ainda, as características microestruturais de cada cerâmica influenciam o comportamento mecânico e adesivo das cerâmicas de dissilicato de lítio e feldspática.

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