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Composite repair: On the fatigue strength of universal adhesives

Thiago Henrique Scarabello Stape[a,b,*], *Oskari Tulkki*[a,b], *Ikram Aqel Salim*[a,b], *Kaveh Nik Jamal*[a], *Mustafa Murat Mutluay*[a,c], *Arzu Tezvergil-Mutluay*[a,b]

[a] *Department of Restorative Dentistry and Cariology, Adhesive Dentistry Research Group, Institute of Dentistry, University of Turku, Turku, Finland*

[b] *Turku University Hospital, TYKS, University of Turku, Turku, Finland*

[c] *Department of Prosthodontics and Clinical Dentistry, Institute of Dentistry, University of Eastern Finland, Kuopio, Finland*

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ABSTRACT

Objectives: To determine whether the composition of universal adhesives and the use of silane coupling agents could affect the fatigue strength of composite repair.

Methods: Composite samples were aged in water at 37 °C for 90 days and bonded to fresh composite to produce twin-bonded bar-shaped composite specimens (2 × 2 × 12 mm). Five universal adhesives, a multistep composite repair system and a hydrophobic solvent-free resin associated to a separate silane coupling agent application were used for bonding. Composite samples were tested under 4-pointflexure initially at quasi-static loading (n = 12) followed by cyclic loading (n = 25). The stress-life fatigue behavior was evaluated following the staircase method at 4 Hz. The unfractured side of cyclic loaded beams were evaluated under SEM to determine crack initiation sites. Fatigue data was analyzed by ANOVA and Tukey test and Wilcoxon Rank Sum Test ($\alpha = 0.05$).

Results: Bonding protocols were unable to restore the cohesive strength of the nanofilled composite ($p < 0.05$). Fatigue testing was more discriminative to reveal discrepancies in composite repair than conventional quasi-static loading. While the composition of universal adhesives affected composite repair potential, the highest endurance limits occurred for the separate silane coupling agent application. Crack propagation sites were mostly located on the aged composite surface.

Significance: Although a trend for simplification invariably overruns current adhesive dentistry, composite repair using solely universal adhesives may result in inferior repair potential. The additonal use of silane coupling agents remains as an important procedure in composite repairs.

* Correspondence to: Institute of Dentistry, University of Turku, Lemminkaisenkatu 2, Turku 20520, Finland.
E-mail addresses: thiago.stape@utu.fi (T.H.S. Stape), oskari.a.tulkki@utu.fi (O. Tulkki), ikram.salim@utu.fi (I.A. Salim), kaveh.nikjamal@utu.fi (K.N. Jamal), mmutluay@utu.fi (M.M. Mutluay), arztez@utu.fi (A. Tezvergil-Mutluay).

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1. Introduction

Methacrylate-based composite restorations are the predominant treatment for cavitated carious lesions in both anterior and posterior teeth. The clinical performance of composite-tooth restorations depends on a myriad number of factors involving patient characteristics, technique used, cavity extension, professional experience and material selection [1–3]. As with any restorative material, composites have a finite lifespan. Although secondary caries is normally considered as the main cause of composite restoration failure [4], fractures may play a bigger role than normally acknowledged on their long-term survival rates [5]. This is believed to be a reflection of the higher use of composites for larger restorations [6]. Annual failure rates varies between 1.9%, in 10–18 years [7], and 2.2%, in 22 years [5]. Higher occlusal stress associated to bruxism and higher caries risk further reduce the lifespan of composite restorations resulting in higher annual failures rates [8,9]. Curiously, caries-related failure rates tend to increase in longer studies, whereas failures associated to composite fracture do not seem to markedly change over time [9]. There is growing evidence that repair increases the longevity of composite restorations [10–13]. Thereby, such minimally invasive procedures have been incorporated into dental school teaching [14–16] and they are gaining popularity among clinicians [17]. Composite repairs are no longer considered as "bad dentistry" [18,19]. In fact, they are characterized as the "contemporary approach" to treat failed restorations [10–12,20,21] not only because of longer restoration longevity, but also due to lower risk of pulp exposure, higher sound tissue preservation, lower treatment time and reduced costs [13,21,22].

Successful composite repair requires good adhesion between the existing aged restoration and the freshly added composite. The main problem in repairing methacrylate-based composite restorations is the reduction, or even absence, of unpolymerized reactive monomers within the surface of the aged restorations. Reactive monomers present various degree of water solubility, depending on their composition, thereby leaching out of the bulk of composite resins in the oral cavity with time. Unavailability of free monomers hinders appropriate covalent bonding between the freshly applied and aged composite through conventional methacrylate free radical polymerization. The ultimate goal in repairing composites is to reestablish their original cohesive strength [23]. Nonetheless, bond strengths between aged and fresh composites vary according to the aging method, rarely reaching the original composite's cohesive strength [23]. Aged composites are less reactive than fresh composites considering interfacial composite–composite bonding ability [23]. Hence, several protocols have been proposed to improve bonding to aged composites [22,23]. They generally consist of increasing physical retention, chemical treatments with silanes or their combination [22,23]. Although there might be controversies [23], an additional silane-application step tends to increase the repair bond strength of methacrylate-based resin composites [20]. To date, a gold-standard protocol has not been established for failed composite restoration [20,22,23].

Recently, silane-containing universal adhesives were introduced with the promise to simplify adhesive dentistry including composite repairs. Monomer composition, solvent type, pH and even silane content vary considerably among universal adhesives [24,25], which potentially affects their bonding performance. Few laboratory studies have evaluated the bonding effectiveness between universal adhesives and aged composites [25] to assess potential differences in composite-composite repair bonding. Therefore, the aim of this study was to evaluate the fatigue strength of composite repairs bonded with different universal adhesives. The effect of a separate pre-hydrolized silane application (*i.e.* not mixed with methacrylate monomers) on composite-composite bonding was also investigated. The central hypothesis examined whether universal bonding agents would produce comparable composite repaired bonded interfaces to the application of a separate silane coupling agent. The tested null hypotheses were that: (i) the composition of commercially available universal adhesives would not affect the fatigue strength of composite repairs and (ii) composite repairs using universal adhesives would not differ from more complex protocols employing separate applications of silane coupling agents.

2. Material and methods

2.1. Experimental design

The fatigue strength of six adhesives systems (Table 1) bonded to aged composite was assessed in terms of quasi-static 4-point flexural strength (n = 12/group), stress life fatigue (n = 25/group) and apparent endurance limit determination. The experimental design was composed of one study factor: adhesive type, in six levels determined by the composition, pH and application mode of each bonding resin. An isolated control group was bonded with a separate silane coupling agent and a solvent-free hydrophobic resin to allow comparisons between different universal adhesives and the recommended composite repair technique bonding technique [22]. Flexural strength, stress-life fatigue behavior and apparent endurance limits were evaluated using the twin-bonded interface (TBI) approach [26].

2.2. Sample preparation and bonding protocols

Bar-shaped composite beams measuring roughly 2 × 2 × 12 mm were produced by layering a nanofilled composite (Filtek Supreme XTE, shade A2B, 3 M ESPE, St. Paul, MN, USA), filler content of 78.5% by weight (63.3% by volume), in a custom-made aluminum mold. Composite increments of 2 mm in thickness were light cured for 20 s using a LED unit (Elipar Deepcure, 3 M ESPE) with tip diameter of 10 mm at 1400 mW/cm^2. Beams were then aged for 90 days in distilled water at 37°C in the dark. Bonding surfaces were wet-roughened with 600-grit SiC (grit size ≈ 16 μm) paper for 20 s to simulate fine-grit diamond burs [22], followed by water-air rinsing and air-drying for 10 s each. Bonding was concomitantly performed on opposing surfaces of the beams placed inside a custom-made aluminum mold. One group

Table 1 – Adhesive systems, composition, classification and application mode.

Adhesive	Composition	~ pH	Classification	Application mode*
Scotchbond Universal (3 M-ESPE; *SU*) *bacth# 3785489*	HEMA; bis-GMA; MDP; ethanol; silane treated silica; water; P205; CAIA; (dimethylamino)ethyl methacrylate; CQ; dimethylaminobenzoat(–4), methyl ethyl ketone	2.7	mild universal	a, b, c, d
Scotchbond Universal Plus (3 M-ESPE; *SU+*) *bacth# 7457878*	2-Propenoic acid, 2-methyl-, diesters with 4,6-dibromo1,3-benzenediol 2-(2-hydroxyethoxy) ethyl 3- hydroxypropyl diethers; 2-HEMA; P205; silane treated silica; ethanol; water; CAIA; N-dimethylbenzocaine; triethoxysilane; acetic acid	2.7	mild universal	a, b, c, d
Clearfil Universal Bond Quick (Kuraray Noritake; *CFU*) *bacth# CL0201*	bis-GMA; ethanol; HEMA; MDP, colloidal silica; silane coupling agent; sodium fluoride; CQ; water	2.3	mild universal	a, e, f, g, b, c, d
Clearfil Repair (Kuraray Noritake; *CFR*) *bacth# 000067*	*K-etchant gel*: Phosphoric acid, water; colloidal silica; dye	< 1	two-step mild self-etch	a, e, f, g, h, c, i, c, d
	Clearfil Porcelain Bond Activator: hydrophobic aromatic dimethacrylate; 3-methacryloxypropyl trimethoxysilane	–		
	Clearfil SE Primer: HEMA; MDP; hydrophilic aliphatic dimethacrylate; colloidal silica; CQ; water; accelerators; dyes	< 2.5		
	Clearfil SE Bond: bis-GMA; 2-HEMA; MDP; hydrophobic aliphatic dimethacrylate; colloidal silica; CQ; accelerators	–		
iBond Universal (Kulzer; *iBond*) *bacth# K010041*	4-META; MDP; methacrylates; acetone; water; CQ; stabilizers	1.7	intermediate strong universal	a, b, c, d
Futurabond M+ (VOCO; *FBM+*) *bacth# 1750404*	HEMA; MDP; bis-GMA; urethanedimethacrylate; ethanol; water, CQ; butylated hydroxytoluene	2.3	mild universal	a, b, c, d
Scothbond Multipurpose (3 M-ESPE; SBMP) *batch # NA20992*	*Bond*: bis-GMA, HEMA, photoinitiators	–	Three-step etch-and-rinse	a, j, b, c, d

Abbreviations: HEMA = Hydroxyethyl methacrylate, bis-GMA = ; MDP = decamethylene dimethacrylate; P205 = 2-Propenoic acid, 2-methyl-reaction products with 1,10- decanediol and phosphorus oxide; CAIA = copolymer of acrylic and itaconic acid; 4-META = 4-methacryloxyethyltrimellitic acid anhydride; CQ = camphoroquinone; *a: roughen (600-grit SiC paper 20 s), rinse and air dry for 10 s; b: adhesive application for 5 s; c: gentle air dry for 10 s; d: light curing for 10 s; e: etchant application (K-etchant gel) for 5 s; f: rinse for 10 s; g: air dry for 10 s; h: silane activation and application for 5 s (mixture of Clearfil SE Primer and Clearfil Porcelain Bond Activator); i: hydrophobic coating application for 5 s (Clearfil SE Bond); j: separate silane application (RelyX Ceramic Primer, 3 M-ESPE) for 60 s. Application modes followed manufacture's recommendations.

was treated for 60 s with a pre-hydrolyzed silane (RelyX Ceramic Primer, 3 M-ESPE), air-dried for 10 s and bonded with a solvent-free hydrophobic resin (Adper Scotchbond Multpurpose, *Bond*, 3 M-ESPE; SBMP). Curing was performed with a LED unit (Elipar Deepcure, 3 M ESPE) at 1400 mW/cm^2. A single operator performed all bonding procedures. Table 1 describes in detail the application modes used for composite repair following manufacturer's recommendations. The nanofilled composite (Filtek Supreme XTE, 3 M ESPE) was subsequently applied in a single increment to fill the mold cavities on both sides of the aged composite beam. Since, surfaces to be cured were larger that the tip of the curing unit, composite surfaces were cured for 20 s on both sides by overlapping. Bonded sections were released from the mold, inspected for voids and flaws using a stereomicroscope at 40 × magnification (Leica M60, Leica Microsystems) and sectioned with a slow-speed diamond saw (IsoMet 1000 Precision Cutter, Buehler Ltd) to obtain composite-to-composite beams, roughly 2 × 2 × 12 mm. Beam surfaces were lightly wet-polished (MetaServ 250 Grinder and Polisher, Buehler) with 600- and 1200-grit SiC grinding paper (Carbimet & Microcut, Buehler Ltd) for 30 s. Two fresh-aged composite interfaces (Fig. 1) were present in each beam forming twin-bonded interfaces (TBI). A minimum of 37 TBI samples were prepared for each group with average cross-sectional area of 3.58 mm^2 (± 0.46). Specimens were re-inspected for flaws at the bonded interface using a stereomicroscope (Leica M60) at 40 × magnification and stored in distilled water at 37°C for 24 h prior to further testing. Samples with detectable flaws were discarded and replaced.

2.3. Characterization of the fatigue behavior

TBI specimens were evaluated under quasi-static and cyclic four-point flexure using a universal testing system (Electropuls E1000, Instron) with load capacity of 250 N and sensitivity of 0.025%. A schematic diagram with the jig, specimen configuration and loading arrangements is shown in Fig. 1. Quasi-static loading was applied at a rate of 0.05 mm/min. The flexural strength (*FS*) of the beams was calculated using conventional beam theory [27] in terms of the maximum measured load (*P*) in N and beam geometry (width *b*, thickness *h* in mm) according to $FS = 3Pl/bh^2$, where *l* is the distance from interior and exterior supports (l = 3 mm). Twelve specimens (n = 12) were evaluated per group. Cyclic loading of the TBI specimens was conducted using the same flexure configuration under load control with frequency of 4 Hz and stress ratio (R = ratio of minimum to maximum cyclic load) of 0.1. The cyclic loading experiments followed the staircase fatigue method and beginning at approximately 95% of the flexural strength, identified from the quasi-static loading, followed by sequential reductions in the order of 10% until failure. The process continued until reaching a flexure stress amplitude (MPa) at which the specimens did not fail within 1.2×10^6 cycles. The cyclic stress amplitude was plotted in terms of the number of cycles to failure in log-base format. The data was fit through a non-linear regression with a Basquin-type model, according to equation $\sigma = A(N)^B$, where A and B are the fatigue-life coefficient and fatigue-life coefficient exponent, respectively. The apparent endurance limit was estimated from the models for a fatigue limit defined at 1×10^7 cycles [26,28]. A minimum of 25 TBI specimens were evaluated per group.

2.4. Detection of crack propagation sites (SEM)

Unfractured sides of TBI specimens that withstood 10^4 loading cycles or more were evaluated by scanning electron microscopy (SEM) to identify the origins of failure and potential weak links at the composite repair bonded interface. Specimens were lightly wet-polished with 600-, 1200-, 2000- and 4000-grit SiC grinding papers (Carbimet & Microcut, Buehler Ltd) for 30 s each, ultrasonically cleaned in distilled water for 60 s, followed by dehydration in silica overnight. Samples were sputtered with gold/palladium and analyzed on backscattering mode at 15 kV (Phenom ProX, PhenomWorld). SEM micrographs (6000 × magnification) were taken sequentially covering the entire extension of the bonded interface at the tensile side of the specimens. A blinded-experienced evaluator identified the most common patterns regarding crack propagation according to the following classification: type I - cracks identified mostly at the aged

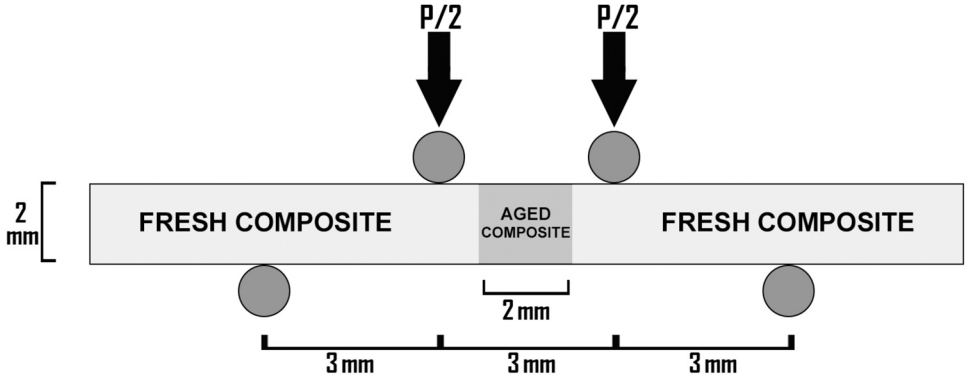

Fig. 1 – Schematic diagram of the twin-bonded interface (TBI) specimen configuration and loading arrangements used for the 4-point-flexure (n = 12), under quasi-static loading, and stress-life fatigue behavior at 4 Hz (n = 25), under cyclic loading. Bonded interfaces were constituted by composite repairs using different intermediate resins and bonding protocols.

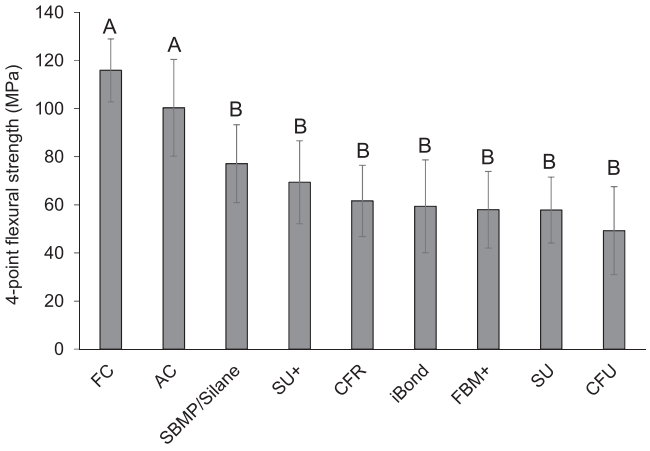

Fig. 2 – Quasi-static loading bond strengths (MPa) for the 4-point bending test at 24 h (n = 12). Composite repair of aged composite (90 days in distilled water at 37 °C) performed with different adhesive systems following the manufacturer's instructions. Fresh (FC) and aged (AC) unrepaired composites served as controls. Bars identified with different capital letters represent significant differences according to Tukey test ($p < 0.05$).

composite-intermediate resin interface or in their proximities; type II - cracks identified mostly at the fresh composite-intermediate resin interface or in their proximities; and type III - cracks identified at both bonded interfaces and within the bulk of the intermediate resin.

2.5. *Statistical analyses*

Data normality and equality of variance of the 4-point flexural strength data set were confirmed by the Shapiro-Wilk and Levene tests, respectively. Flexure strengths obtained after quasi-static loading measurements were analyzed using a one-way ANOVA and Tukey test. Fatigue life distribution data was analyzed using the Wilcoxon Rank Sum Test. Significance levels were set at 5% ($\alpha = 0.05$). Statistical analyzes were performed on IBM SPSS Statistics for Windows, version 23 (IBM Corp., Armonk, NY, USA).

3. Results

3.1. *4-point flexural strength*

TBI specimens (overall mean cross-sectional and standard deviation: 3.52 $mm^2 \pm 0.41$) presented no significant differences regarding specimen size between groups ($p = 0.762$). Composite cohesive strength and bond strengths of composite repaired interfaces are shown in Fig. 2. One-way ANOVA revealed that the tested bonding protocols produced no significant differences on composite-composite 4-point flexural strength. Bonding protocols were unable to restore the flexural strength of the nanofilled composite, producing flexural strengths 34–55% lower. Water storage for 90 days had no significant effects on the nanofilled composite flexural strength. All fractures involved the bonded interface; no pure cohesive fractures were observed in neither aged nor fresh composite samples. Fractures originated at the tensile side of all bar-shaped specimens.

3.2. *Fatigue behavior and resistance*

Fatigue life diagrams (S-N curves) for the TBI specimens are shown in Fig. 3. Regression analyses with basquin-type power law models are listed for all groups to describe the mean fatigue strength distribution. Wilcoxon Rank Sum test revealed significant differences in stress amplitude between groups. Table 2 shows pairwise comparisons between fatigue life distributions adjusted by Bonferroni correction for multiple tests. The highest fatigue strength was observed for the unrepaired fresh and aged composites without significant differences between them. All bonding protocols produced significantly lower fatigue strengths than the unrepaired composites. Stress amplitudes of composite repairs using a separate silane coupling agent followed by the application of a hydrophobic bonding resin were significantly higher than those produced by the universal adhesives. No significant differences occurred between universal adhesives with the exception of CFU, which produced significantly lower stress amplitudes. The stress-life fatigue constants were used to estimate the apparent endurance limit for all groups at 1×10^7 cycles and they are listed in Table 2. The highest apparent endurance limits for composite repair occurred when a separate silane coupling agent (SBMP + Silane) was used. Clear discrepancies between universal adhesives were identified regarding their endurance limits. CFR and SU+ produced the highest endurance limits for the universal adhesives, which were 68% and 20% higher than CFU and SU, respectively. FBM+ and iBond produced apparent endurance limits 40% and 21% lower than CFR, respectively.

3.3. *Crack propagation sites*

The overall crack propagation sites at bonded interfaces varied according to bonding protocols. Crack propagation sites were observed in all evaluated samples. Although crack formation was observed on both fresh (type II) and aged (type I) composite interfaces, they were mostly located on the latter. This depicts the poorer interaction between intermediate bonding resins and aged composites (Fig. 4), regardless of their composition. Hence, type I, II and III patterns were observed in all groups; however, differences in their incidences were clearly observed. In fact, the most prevalent pattern observed in each group corresponded to at least 84% of crack propagation sites for that specific bonding protocol. SBMP + Silane interfaces were mostly characterized by the type I pattern, but with the lowest number of detectable crack propagation sites at the aged composite and bonding resin. CFR and SU+ interfaces presented mostly the type I pattern with similar crack distribution patterns and fewer detectable cracks than the remaining universal adhesives. iBond interfaces presented mostly the type I pattern with the lowest number of crack propagation sites at the fresh composite and bonding resin considering only universal adhesives. The aged composite-iBond interface presented similar crack distribution as CFR, SU+ and SU, but with higher loss of filler

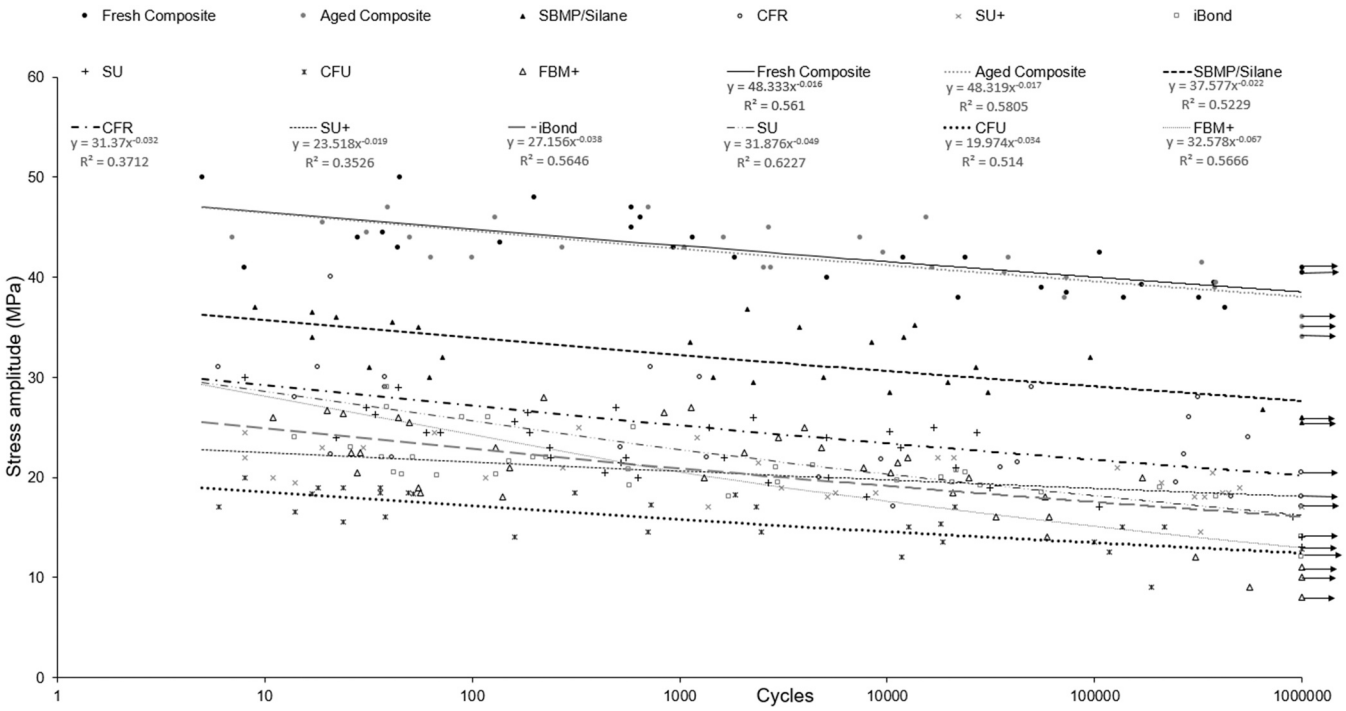

Fig. 3 – Stress life diagrams (S-N curves) for composite repair of aged composite performed with different adhesive systems. Note that data points with arrows represent specimens that reached 1.2×10^6 cycles and the test was discontinued. R^2 values represent the coefficient of determination.

Table 2 – Stress-life fatigue response, power law constants, estimated endurance limits and pairwise comparisons for composite-repair interfaces bonded with different universal adhesives.

	A (MPa)	B	R^2	Endurance Limit (MPa)	Endurance limit reduction[a] (%)	Adjusted Significance[b]
Fresh composite	48.333	-0.016	0.56	36.7	–	a
Aged composite	48.319	-0.017	0.58	36.6	-0.3	a
SBMP/Silane	37.577	-0.022	0.52	26.4	-28.2	b
CFR	31.370	-0.032	0.37	18.7	-49	c
SU+	23.518	-0.019	0.35	17.3	-52.9	c
iBond	27.156	-0.038	0.56	14.7	-59.9	c
SU	31.876	-0.049	0.62	14.5	-60.1	c
FBM+	32.578	-0.067	0.57	11.1	-69.9	c
CFU	19.974	-0.034	0.51	11.6	-68.6	d

R^2 values represent the coefficient of determination for each model. Endurance limits were calculated at 1×10^7 cycles. [a] Fresh composite used as baseline reference. [b] Pairwise comparisons between fatigue life distributions adjusted by Bonferroni correction for multiple tests.
Abbreviations: SBMP = Scotchbond Multporpose, Bond, 3 M-ESPE; SU = Scotchbond Universal, 3 M ESPE; SU+ = Scotchbond Universal Plus, 3 M ESPE; CFU = Clearfil Universal Bond Quick, Kuraray Noritake; CFR = Clearfil Repair, Kuraray Noritake; iBond = iBond Universal, Kulzer; FBM+ = Futurabond M+ , VOCO.

particles. SU repaired interfaces presented mostly the type I pattern with higher distribution of cracks at the aged composite interface than CFR, SU+, SU and iBond. The highest incidence of crack propagation sites was observed for FBM+ and CFU presenting mostly the type III pattern. Crack distribution did not differ between. FBM+ and CFU.

4. Discussion

Replacement of failed restorations in dentistry is normally costly and time consuming. As composite repair gains popularity in the daily clinical life of dentists, determining the most efficient bonding technique to produce strong links between the aged and freshly applied composite becomes necessary. This could greatly affect the longevity of composite restorations and reduce reservice times. Since the composition of universal adhesives affected fatigue strengths, represented by the apparent endurance limits of composite repairs, the first null hypothesis was rejected.

Different methods have been used to evaluate the bonding performance of composite repairs including the so-called shear/microshear [29,30], tensile/microtensile [31–33] and interfacial fracture toughness tests [34]. To date, there is a

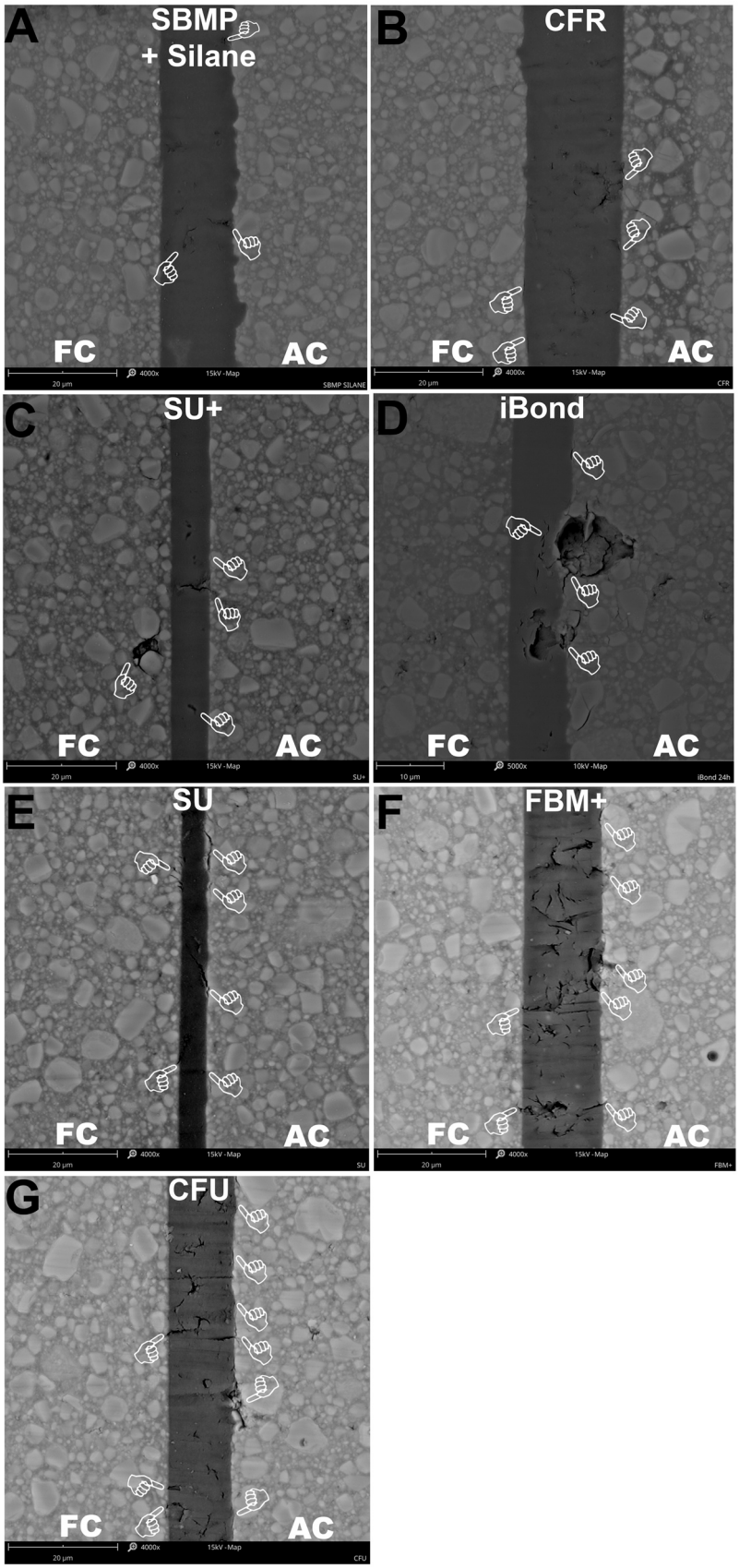

Fig. 4 – Representative SEM micrographs of composite repaired specimens bonded with different universal adhesives. Profile view of TBI specimens (unfractured interface) subjected to a minimum of 10^4 cycles. Pointing fingers show examples of crack propagations. Abbreviations: FC = Fresh composite; AC = Aged composite.

predominance of shear/microshear testing for composite repair [20,23]. Altogether, variations in test setups and mechanics certainly contribute to the conflicting-inconclusive findings currently present in the dental literature regarding composite-composite bonding [23]. While the shear/microshear test is relatively simple and easy to perform, inadequate stress distributions may produce questionable outcomes due to the great number of cohesive failures. Furthermore, "pure" shear stresses are not easily produced with conventional microshear test setups used in dental research [35]. In this scenario, failures comprise areas with higher stress concentration and not necessarily correspond to the weakest portions of the bonded interface. Hence, microshear cannot be characterized as the most reliable method to test such composite-composite interfaces [35,36]. Differently, microtensile testing generates more uniform stress distribution along the bonded interface and it is usually the first choice of bond strength testing when applicable. Composites are not as brittle materials (*i.e.* compared to enamel), so flaws produced during sample sectioning should not be impeditive to subject them to microtensile testing. Microtensile testing is considered a more reliable approach than microshear testing for composite repair assessments [37]. Nonetheless, stress distribution is still not ideal, which raises concerns about the true maximum stress bonded interfaces resist at fracture [35,38]. The recently proposed micro-interfacial fracture toughness can be considered as a more accurate approach to reveal interfacial properties being less test dependent [39]. However, it still relies exclusively on monotonic loading to determine bonding effectiveness. In adhesive dentistry, quasi-static loading is a common trait between current test methods (*i.e.* microshear and microtensile used to determine interfacial bonding effectiveness). In such setups, failures are generally associated with overloads and not necessarily due to lower strain cyclic loading. Typical masticatory stress levels that a human tooth experiences are in the order of 20 – 42 MPa [40]. Under normal physiological conditions, composite failures take place at considerably lower cyclic loads before reaching the high stresses at the bonded interfaces observed during monotonic testing (*i.e.*,~ 50 – 70 MPa). Acknowledging only monotonic values may lead to false implications regarding composite repair bonding potential. The ability of test methods to discriminate between bonding protocols is critical when it comes to determining the best performing protocols. Controversies found in previously reported composite-repair findings [20,29,37,41,42] may thus be at least partially attributed to the lower discriminative power of monotonic test setups, which do not necessarily take place in a consistent-reproducible manner. Contrary, fatigue testing of twin-bonded interfaces following the staircase approach, has been shown to be more discriminative and reliable than conventional monotonic quasi-static loading [43]. To the best of our knowledge, repaired composite-composite interfaces have not been tested under cyclic loading. Thereby, this study employed a fatigue-testing approach to help elucidate possible method-related inconsistencies on the use of intermediate bonding resins for composite repair. The present findings characterize an original insight regarding the fatigue behavior of composite repaired interfaces. For instance, if only the 4-point flexural monotonic setup were employed, the superior performance of a separate silane application would not be identified. The tested universal adhesives would also be categorized with similar composite repair potential, when in fact, their ability to bond to water-aged composite varied. This emphasizes the importance of employing adequate methods, including cyclic loading setups, for testing bonded interfaces to avoid possible generalizations that may mislead assumptions regarding bonding protocol selection in dentistry.

In order to investigate *in vitro* composite-composite bonding, proper composite aging before repair is crucial to determine their true effectiveness. Water storage is the method most commonly used in composite-repair studies, albeit low storage times have been routinely employed [23]. Short-term water storage (below 7 days) can be considered inadequate [23] since overestimation of the repair potential is likely to occur on account of residual reactive monomers that contribute to additional chemical bonding. The presence of unreacted vinyl groups (C=C) is unlikely to take place at the surface of intra-oral aged composites after a few months of clinical service. Typical resin-based dental restorations become saturated with water within only one to two months after placement [44]. The rationale for selecting static storage in water at 37 °C for 90 days was that, given such time, near-maximum water uptake and polymer solubility is established. This certainly maximized elution of reactive-monomer out of the composite surface. Restorations are normally prone to be repaired in the medium or long-term clinical service [1–3,45] reinforcing the necessity of longer *in vitro* water storage before repairs to better simulate more realistic bonding scenarios. The ultimate goal of composite repairs is to match the original composite cohesive strength. In theory, this would represent full repair potential. It has been reported that matching the original composite strength is possible by employing intermediate bonding resins [32,46]; however, our findings do not support the latter especially when a composite with relatively high filler content is used. Most studies claiming equivalent repair potentials to composite cohesive strength employed short-term water storage for up to 7 days [23]. Hence, overestimated composite repair values were likely to happen. In the present study, both quasi-static 4-point flexural strengths and fatigue strength measurements indicate that restoring the original cohesive strength of aged composites using only intermediate resins, with or without a separate silane application, is unlikely to occur. The high incidence of crack propagation sites at the aged composite characterizes such less reactive interface as the weakest link in composite repair. This reinforces the necessity to improve the interaction between aged and fresh composites in order to produce stronger bonded interfaces. Considering the best tested scenario (*i.e.* SBMP/Silane), flexural bond strengths were still 33 % lower than the composite cohesive strength. Apparent endurance limits of composite-repair interfaces also pointed out in the same direction showing low values, – 28 % compared to the unrepaired nanofilled composite. Although full composite repair potential with current universal adhesives is uncertain considering composites with a relatively high filler content, identification of the best-performing bonding protocols could shed some light in this conflicting topic and contribute to more favorable

evidence-based decisions. It is evident that the chemical composition of universal adhesives affected composite repair at least to some extent. This would certainly go unnoticed if only monotonic tests were employed, confirming the higher discriminative power of fatigue test setups.

Composite repair strength can be considered adhesive-composite dependent [34]. Composite repairs rely on micromechanical retention and/or additional chemical bonding to composite fillers. Hence, surface roughening is an important step [22]. It creates microretention and also exposing filler particles that participate in chemical adhesive-composite bonding. Considering the available roughening methods, alumina sandblasting tends to produce higher composite repair bond strengths than bur roughening [47], albeit both approaches seem to be equally effective for the tested nanofilled composite [48]. The rationale for only roughening composite surfaces with 600-grit SiC paper was to simulate the abrasion of fine-grit diamond burs, which has greater clinical applicability and acceptability by clinicians. Surface roughing exposes not only silica, but also zirconia fillers, which ionically bond to MDP is a concentration dependent manner [49]. MDP is currently present in most universal adhesives and may improve repair bond strengths to zirconia-based composites. Unfortunately, determining the specific extent in which MDP content in adhesives affected fatigue strength is virtually impossible due to manufacture's secrecy on adhesive formulations. It is not possible to know the specific MDP content in each bonding resin to make realistic assumptions. We can only speculate that fatigue strengths reported here might not be extrapolated to zirconia-free composites. The lack of MDP-zirconia chemical bonding may produce inferior outcomes. Differently, the effect of silane content on fatigue strength could be verified.

The experimental design deliberately included two silane-free universal adhesives (iBond and FBM+), to determine whether the incorporation of silanes into universal adhesives improves repair potential especially for commonly used composites with high filler content. Since the tested silane-free universal adhesive produced flexural bond strengths that were not significantly different from the silane-containing bonding resins, it is possible to imply that silane incorporation into universal adhesives does not strongly benefit composite-composite bonding when samples are properly aged before bonding. Moreover, the lack of silane incorporation did not produced significantly lower stress amplitudes among most silane-containing bonding resins. Curiously, iBond outperformed one of the silane-containing adhesives (CFU). This reinforces the assumption that silane incorporation into universal adhesives may not be as relevant in repairing highly filled composites as commonly believed . The inability of silane-containing universal adhesives to effectively improve composite repair bond strengths has been previously proposed [20,41]. Silanes in acidic conditions may become unstable due to the self-condensation reaction of silanol groups. Methacrylate monomers may also interfere with the silane-coupling condensation reaction between silanes and hydroxyl groups (-OH) found in silica fillers [50,51]. Even though flexural strengths and isolated stress amplitudes suggest that silane incorporation into universal adhesives has no effect on composite repair, it is important to note that static evaluations may not be reliable predictors of bonding performance [43]. A clearer view of the composite repair potential of universal adhesives is possible by examining their fatigue strength through their apparent endurance limits. For instance, CFR (18.73 MPa) stood out as the best performing composite repair bonding resin, which contained silane coupling agent mixed with acidic monomers. CFR was closely followed by SU+ (17.31 MPa). The apparent endurance limit of CFR was 28 % higher than iBond (14.72 MPa), the third highest, and 70 % higher than FBM+ (11.06 MPa), the lowest endurance limit. Therefore, it becomes evident that the composition of universal adhesives indeed affected composite repair. Although CFR incorporates a silane application step, where the silane is mixed with methacrylate monomers, the lower silane-monomer ratio compared to the tested universal adhesive likely minimized the negative effect of monomers on the ability of silane coupling agents to chemically bond to the silica fillers. Moreover, formulation of new silane compounds, such as the one recently released in SU+ , may be a viable alternative to circumvent some of the limitations associated to silane-monomer combinations when simplification of bonding procedures must be prioritized.

In the context of striving for the optimal composite repair potential, separate silane application produced the best outcomes, so the second null hypothesis was rejected. Higher endurance limits, in the order of 40 %, and also significantly higher stress amplitudes than the best performing universal adhesive reinforce the importance of a separate silane application in composite repair. This is supported by previous studies highlighting the benefits of using silane coupling agents in composite repair [20,41]. The absence of methacrylate monomers during silanization seems necessary to benefit composite repairs [50,51] making the added bonding step worth the extra time. Considering the hydrolytic cleavage of the siloxane bond [52] over time, further studies are necessary to investigate the fatigue strength of composite repaired interfaces after long-term water storage. Normally, filler loading into methacrylate-based dental composites varies between 50 % and 85 % by weight and 35 – 70 % by volume [53]. Since the tested composite presents a relatively high filler content (*i.e.*, 78.5 % and 63.3 % by weight and volume, respectively), one limitation in the present study was the absence of composites with lower filler content. In such conditions, the effect of silane on the composite repair fatigue strength may be reduced. Future studies should thereby include, composites with different filler contents and monomer compositions. Furthermore, future improvements in silane formulations designed for incorporation into resin blends are required to potentially match the boding effectiveness of a separate silane application.

5. Conclusion

Within the limitations of this study, restoring the cohesive strength of composites with relatively high filler content *via* repair using solely intermediate bonding resins was not possible. While the conventional 4-point flexural monotonic

test was unable to identify differences between the various universal adhesives, the fatigue-testing approach was more discriminative to reveal discrepancies in composite repair potential. The composition of universal adhesives was a determinant factor on composite repair potential, indicating that selection of universal adhesives is critical to produce interfaces with improved fatigue strength.The use of a separate silane coupling agent remains as an important boding step to improve the repair potential of composites with relatively high filler content.

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