ORIGINAL ARTICLE

The competition between enamel and dentin adhesion within a cavity: An in vitro evaluation of class V restorations

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Abstract To gain more insight into the consequences of curing contraction within the tooth cavity, we assessed the margin behavior of 12 contemporary restorative systems in class V restorations with margins located on enamel and dentin after mechanical loading and water storage. Mixed class V cavities were prepared on extracted human molars and restored using five etch and rinse and seven self-etch adhesive systems with their corresponding composites. Marginal adaptation was evaluated by using a computer-assisted quantitative marginal analysis in a scanning electron microscope (SEM) on epoxy replicas before, after thermal and mechanical stressing and after 1 year of water storage. The interactions of "testing conditions", "adhesive–composite combination" and "tooth substrate" with "marginal adaptation" were evaluated by two-way ANOVA. Fatigue, stress and storage conditions had significant effects on the marginal adaptation. Only two groups (Optibond FL and G Bond) presented equal percentages of marginal adaptation on enamel and dentin; in the other groups, the rate of degradation was product dependent. All materials tested showed a distinct behavior on enamel and dentin. In addition to mechanical resistance and long-term stability, differences within materials also exist in their ability to simultaneously bond to enamel and dentin.

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Introduction

Direct adhesive fillings are progressively becoming the restorative procedure of choice in modern operative dentistry [1, 2]. One of the requirements for this type of restoration is to achieve a stress-resistant adhesion between tooth substrate and filling composite to ensure restorations' marginal integrity and retention. It has been suggested that degradation of the resin–dentin interface basically occurs in three steps: water is absorbed into the polymer component of the adhesive system, then resin is eluted from the hybrid or adhesive layer and finally, exposed collagen fibrils are degraded by matrix metalloproteinase coming from either dentinal fluid or saliva [3–7].

Enamel preservation at the restoration margins plays an important role in the protection of the resin–dentin interface against degradation. If dentin surfaces are protected by peripheral enamel, the resin–dentin interface can better resist chemical attack [8–14]. In terms of vectors generated by curing contraction of the restorative composite, it is known from early studies [15, 16] that in the case of restorations with margins located on enamel and floors cut into dentin, the use of bonding agents that cannot withstand composites' contraction stress will result in detachments from the dentin part of the cavity while the composite restoration will remain attached only to the enamel walls. However, many cavities in the clinical situation, especially the ones located in the cervical area, can involve both enamel and dentin margins. In this context, a recent study has found an important effect of the tooth substrate (enamel and dentin) on adhesion [17]. The authors observed that

within a restoration–adhesive interface, there are stronger and weaker areas of attachment to the tooth tissue. This means that if contraction stresses due to polymerization overcome the weakest attachment to the tooth, the restoration surface will be detached from this area and will shrink to the area that offers the most durable adhesion or towards the intact bonding areas. They concluded that the bonding quality at the interface restoration–tooth is a critical factor for evaluating the direction of polymerization contraction. However, this study reported the results in terms of deformation analysis of the resin composite, but no effect on marginal adaptation was investigated.

The main parameters evaluated for rating the clinical success of a restoration are: retention or percentage of lost restorations, marginal adaptation, color match, marginal discoloration, secondary caries and surface roughness. The percentage of lost restorations is considered an important parameter in the material's bonding ability. However, microleakage and not retention, has been reported to be the primary cause of clinical failure in cervical restorations [18]. Because microleakage occurs at the composite–tooth interface, initial signs of adhesive degradation can be already visible at the restoration margins before the restoration detaches from the tooth cavity. Dental restorations are continuously subjected to several environmental factors such as the presence of moisture and saliva, chewing forces, changes in temperature and pH, and chemical and enzymatic attack. Similar to dentin, enamel adhesion could be equally prone to degradation over time and deformation vectors generated by curing contraction [17] could influence the adhesive interface's integrity at the marignal level.

Therefore, it was the purpose of this in vitro study to investigate whether 12 contemporary adhesive–composite combinations are able to provide a stress-resistant adhesion on enamel and dentin after the effect of artificial aging conditions.

The null hypothesis tested was that there would be no difference in marginal integrity and degradation potential of enamel and dentin with the different materials tested.

Materials and method

Selection and preparation of teeth

The setup of the study is resumed in Fig. 1. Caries-free human molars stored in 0.1% thymol solution at 9°C were used for the experiment within a month after extraction. After scaling and pumicing, the teeth were mounted on custom-made specimen holders with their roots in the center using a cold-polymerizing resin (Technovit 4071,
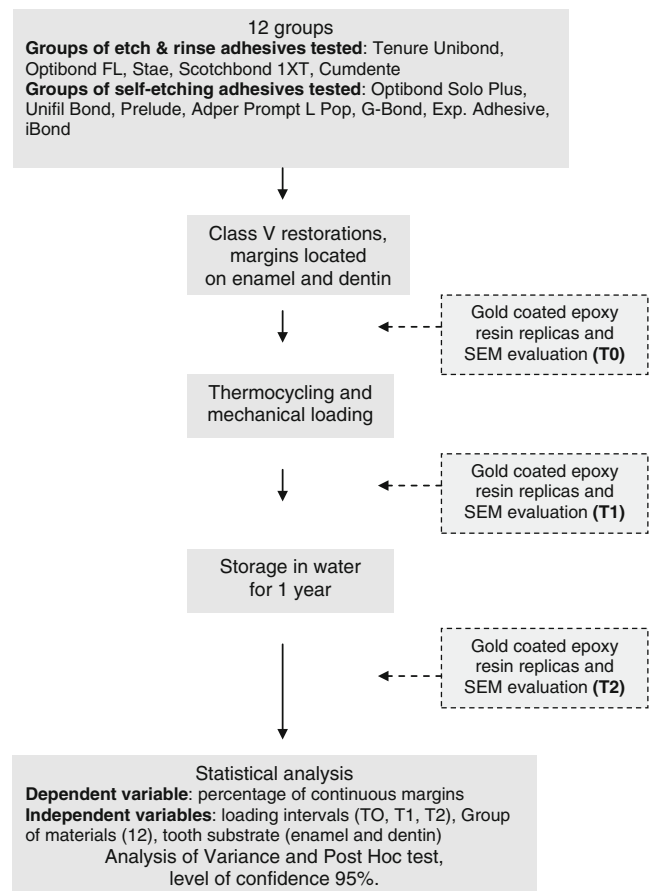

Fig. 1 Set up of the study

Heraeus Kulzer GmbH, Wehrheim, Germany) and then randomly assigned to 12 experimental groups (Table 1). Prior to the mounting procedure, the apices were sealed with two coats of nail varnish. To simulate dentinal fluid flow, a cylindrical hole was drilled into the pulpal chamber approximately in the middle third of the root and a metal tube, with a diameter of 1.4 mm, was then adhesively luted using a dentinal adhesive (Syntac Classic, IvoclarVivadent AG, Schaan, Liechtenstein). The pulpal tissue was not removed. This tube was connected by a flexible silicone hose to an infusion bottle placed 34 cm vertically above the test tooth. The infusion bottle was filled with horse serum (PAA Laboratories GmbH, Linz, Austria) and phosphate-buffered saline solution (PBS; Oxoid Ltd., Basingstoke, Hampshire, England) diluted in a 1:3 ratio under a hydrostatic pressure of about 25 mm Hg. Twenty-four hours before starting the cavity preparations, dentinal fluid was evacuated through the pulp chamber with a vacuum pump by using a three-way valve and subsequently bubble-free filled with the above solution. As of this moment, the intrapulpal pressure was maintained at 25 mm Hg through-out the testing, i.e., during cavity preparation, restoration placement, finishing and stressing.

Table 1 Description of the experimental groups

| Class of adhesive (number of components/ number of steps)	Groups *adhesive system* filling composite	Manufacturer	Batch number
Etch and rinse (4c/3 s)	*Tenure UniBond* Virtuoso	Den Mat, Santa Maria, California, USA	35% H_3PO_4, Solution A 039851901, Solution B 039851902, Gloss 030451001, Filling composite 030381953
(3c/3 s)	*Optibond FL* Premise	Kerr, Orange, CA, USA	35% H_3PO_4, primer 414998, bond 014550, filling composite 011572
(2c/2 s)	*Stae* Ice	SDI, Bayswater, Victoria, Australia	37% H_3PO_4 03071120, adhesive 030720, filling composite 030831
	Scotchbond 1XT Filtek Supreme	3 M ESPE, Seefeld, Germany	35% H_3PO_4 4CF, adhesive 4AJ, filling composite AM
	Cumdente Viscous	Cumdente GmbH, Tübingen,Germany	35% H_3PO_4, adhesive 5409429, filling composite 301547
Self-etch (2c/2 s)	*Optibond Solo Plus* Premise	Kerr, Orange, CA, USA	Primer 408740, bond 408200, filling composite PR-A2
	UniFil Bond Gradia	GC Corporation, Tokio, Japan	Primer 0309021, bond 0309011, filling composite 0302241
	Prelude Accolade	Danville, California, USA	Primer 6763, bond 6764, filling composite 6684
(2c/1 s) (1c/1 s)	*Adp Prompt L Pop* Filtek Supreme	3 M ESPE, Seefeld, Germany	Single component L5 198902, Filling composite 5FG
	G Bond Gradia	GC Corporation, Tokio, Japan	Single component 0404011 Filling composite 0302241
	Exp. adhesive Clearfil AP-X	Kuraray Medical Inc., Tokio, Japan	Single component 040219 Filling composite 00824B
	iBond Venus	Heraeus Kulzer, Dormagen, Germany	Single component 010046 Filling composite 030023

c components, *s* steps

Cavity preparation

Seventy-two V-shaped standardized class V cavities were prepared on the teeth's cervical area [19] with half of the margins located in enamel, half in dentin (Fig. 2) and further restored with the different resin composites detailed in Table 1. Eighty micrometer diamond burs (Diatech Dental, Coltène-Whaledent, Altstätten, Switzerland) were used under continuous water cooling; each bur was replaced by a new one after three cavity preparations. The dimensions of the V-shaped cavities were measured with a periodontal probe and their size was of 3.0–3.5 mm in mesiodistal direction, 2.5–3.0 mm in occlusogingival direction and 1.5 mm in depth. The margin in enamel was bevelled to a crescent shape with a maximum width of 1.2 mm. The entire cavity was finished using 15 μm finishing diamond burs (Diatech Dental, Coltène-Whaledent, Altstätten, Switzerland). Then, the cavity preparations were checked for marginal imperfections such as fractures or chipping under an optical microscope (Wild M5, Wild AG, Heerbrugg, Switzerland) at 12× magnification and corrected if necessary.

Restoration placement

The adhesive systems and their corresponding restorative composites were applied following the manufacturers' recommendations. One operator prepared, restored and polished the restorations. This operator could not be blinded as during cavity filling, application instructions had to be carefully read before application of each product. After the placement and light-curing of the adhesive system by using a halogen light source (Optilux 501, Kerr/Demetron, Danbury, CT, USA) with a constant relative power density output of at least 800 mW/cm^2 (Curing Radiometer Model 100, Serial No. 134089, Demetron Research Corp. Danbury, CT, USA), the composites were inserted into the cavity in two layers, the first layer being placed cervically up to one-half of the cavity and the second layer occlusally filling the other half of the cavity. Both layers were light-cured for 40 s each (Optilux 501). Immediately after polymerization, the restorations were finished and polished by using flexible aluminium oxide discs with different grain sizes (SofLex PopOn, 3 M ESPE AG, Seefeld, Germany). The final polishing was controlled using an optical

Fig. 2 SEM micrograph (50× magnification). Representative frontal view of a class V restoration with margins located on enamel and on dentin. A gap can be observed in several segments of the dentinal margin while no gaps are visible on enamel margins
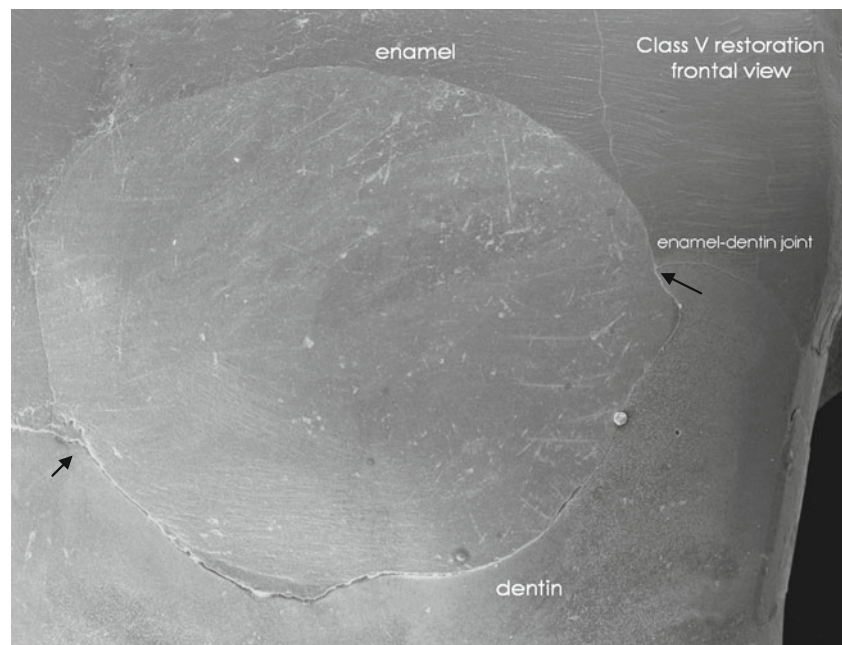
microscope under 12× magnification and corrected if necessary.

Thermomechanical loading

After storage in the dark in 0.9% saline solution at 37°C for 1 week, the restored teeth were loaded in a computer-controlled chewing machine [20]. Thermal and mechanical loading were applied simultaneously. Thermal cycling was carried out in flushing water with temperatures changing 3,000× from 5°C to 50°C with a dwelling time of 2 min each. The mechanical stress comprised in total 1.2 million load cycles transferred to the center of the occlusal surface with a frequency of 1.7 Hz and a maximal load of 49 N applied by using a natural lingual cusp taken from an extracted human molar. Simulation of dentinal fluid flow was permanently maintained throughout the loading procedure.

Water storage

Following the loading procedure, the teeth were stored in 0.5% chloramine-containing water to prevent bacterial growth [21]. Throughout the 12-month immersion period, the teeth were placed in a Memmert oven (Schwabach, Germany) at a constant temperature of 37°C.

Assessment of marginal adaptation

Immediately after completion of the polishing procedure (T0), after loading (T1), and after loading and 1 year water storage (T2), the teeth were cleaned with rotating brushes

and toothpaste. Then, impressions with a polyvinylsiloxane material (President light body, Coltène-Whaledent, Altstätten, Switzerland) were made of each restoration. Subsequently, gold-coated epoxy replicas were prepared for the computer-assisted quantitative margin analysis in a scanning electron microscope (XL20, Philips, Eindhoven, The Netherlands) at 200× magnification by using a custom-made module programmed with an image processing software (Scion Image, Scion Corp., Frederik, MA 21703, USA). All specimens were subjected to the quantitative evaluation, examined by a blinded and trained lab technician, and classified according to the criteria detailed in Table 2. The marginal quality, expressed in percentages of continuous margins

Table 2 Criteria used for the quantitative margin evaluation

Microscopic quantitative evaluation of restoration margins	
Margin type	Explanation
Percentage of continuous margins	• No gap, no interruption of continuity
Percentage of noncontinuous margins	• Gap due to adhesive or cohesive failure
	• Fracture of restorative material
	• Fracture of enamel related to restoration margins
Composite overhangs	• Excess of material at the restoration margins
Underfilled margins	• Margins not covered by composite resin

(% CM), was reported for the total marginal length (average value of enamel and dentin marginal adaptation), as well as for enamel and dentin margins separately at each interval *T*0, *T*1 and *T*2.

Statistical analysis

Statistical analysis was performed with SPSS 14.0 for Windows. Levene's test was used to assess the equality of variance in the different samples. In fact, this procedure tests the null hypothesis that variances of the populations from which different samples are drawn are equal. As the *p*-value was higher than 0.05 (*p*=0.06), the null hypothesis could not be rejected; therefore, we assumed equality of variances. Kolmogorov–Smirnov tested the null hypothesis that the samples came from a normally distributed population. As the resulted *p*-value was higher than 0.05 (*p*=0.079), this null hypothesis could not be rejected; therefore, the data was normally distributed. Equality of variances of samples and normally distributed data enabled the use of a two-way analysis of variance (ANOVA) to study the effects of: 1. *Testing intervals* (*T*0, *T*1, and *T*2) and *adhesive–composite* combinations (the 12 groups) on the marginal adaptation and 2. *Tooth substrate* (enamel and dentin) and *adhesive–composite* combinations on the marginal adaptation. Duncan post hoc test was used to visualize differences in marginal adaptation means among groups. The confidence level was set to 95%. To determine if the number of samples was adequate, group size was statistically evaluated with specific software (Statistics Calculator, StatPac, Inc., Bloomington, MN, USA). The software asked to introduce three values: 1. The population standard deviation (in the case of our study, the mean standard deviation was around 12.8), 2. The maximum acceptable difference, i.e., the maximum difference that the sample can deviate from the true population mean before one can call the difference "significant" (in the present study, a maximum deviation of ten was considered acceptable) and 3. The confidence level that was established as 95%. The sample size given by the calculator was of six, as used in the present study.

Results

The results of marginal adaptation, presented as the mean value of *T*0+*T*1+*T*2, for the *total margin length*, *enamel* and *dentin* margins are detailed in Figs. 3 and 4. The highest scores of marginal integrity, when the *total margin length* was considered, were observed in a three-step etch-and-rinse (Fig. 3a: Optibond FL, % CM of 73.7±12.6) and a one-step self-etching adhesive (Fig. 4a: G Bond, % CM of 78.8±8.1).
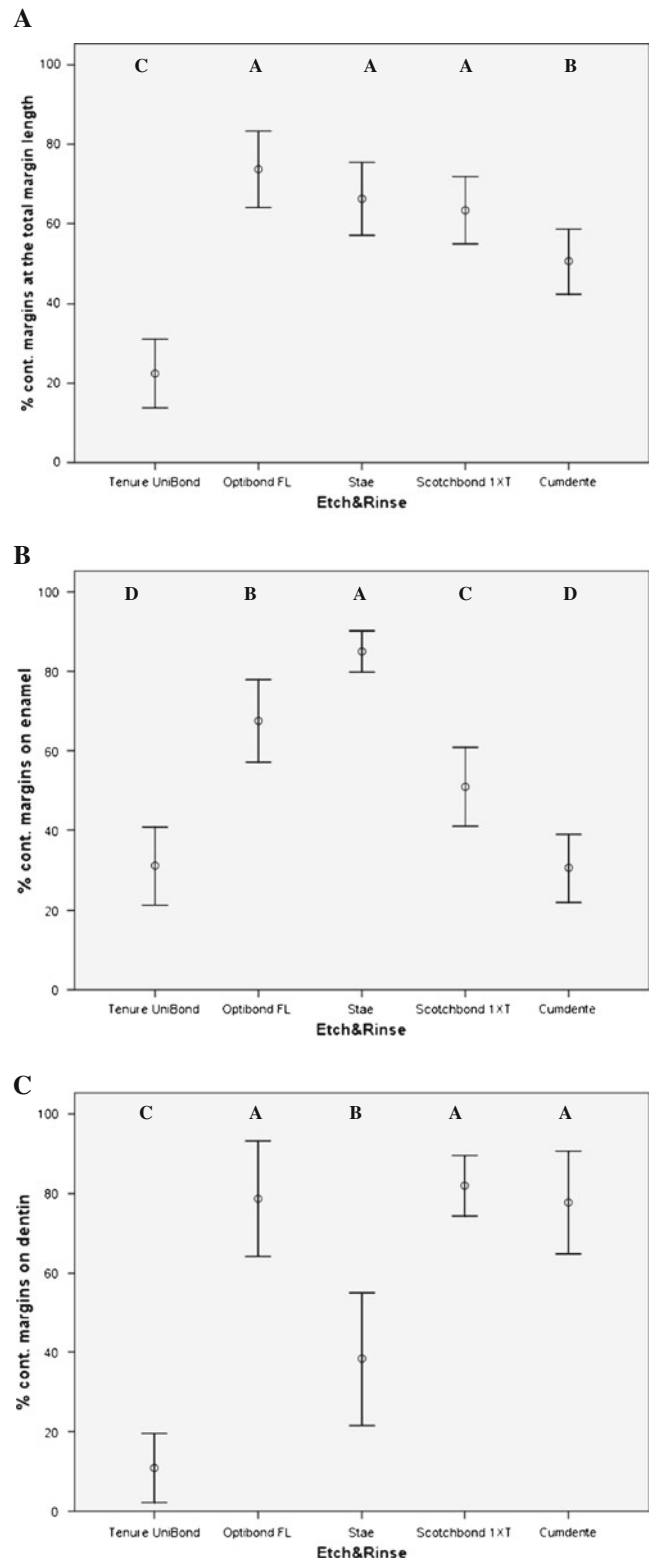

Fig. 3 Etch and rinse groups. Graphic representation of the percentage of continuous margins after testing observed on the total margin length (**a**), on enamel (**b**) and on dentin (**c**)

In the case of the etch and rinse groups, no significant differences at the *total margin length* could be detected
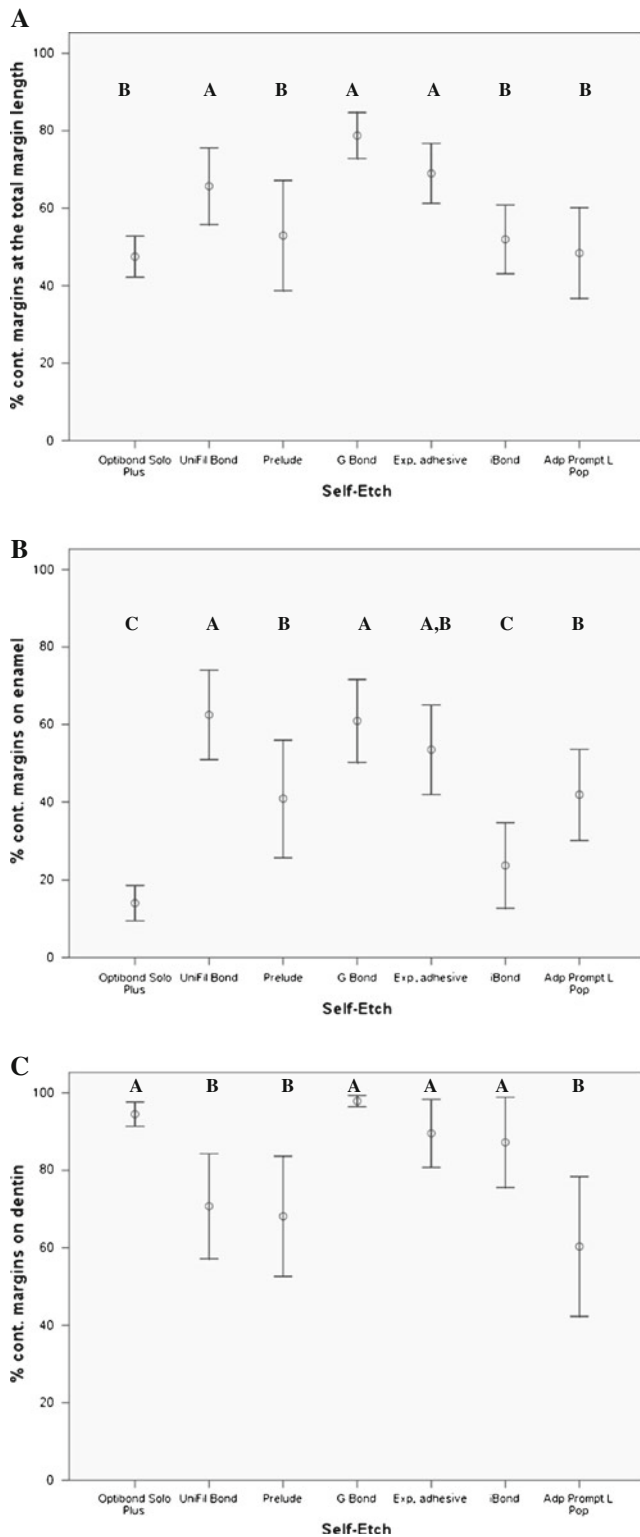
Fig. 4 Self-etch groups. Graphic representation of the percentage of continuous margins after testing observed on the total margin length (**a**), on enamel (**b**) and on dentin (**c**)

between Optibond FL, Stae and Scotchbond 1XT (Fig. 3a, groups connected by the letter A). However, when *enamel* and *dentin* margins were evaluated separately (Fig. 3b and c),

a distinct behavior was observed in these groups with respect to both tooth substrates. For example, Stae delivered more continuous or gap-free margins on enamel (Fig. 3b: % cont. margins 85±10.4) than on dentin (Fig. 3c: 38.3±33.8). Scotchbond 1XT was the contrary; a higher percentage of continuous margins was observed on dentin (Fig. 3c: 82± 15.2) with respect to enamel (Fig. 3b: 50.9±20). Optibond FL was the only material that presented a relatively equal marginal adaptation on enamel (Fig: 3b: 67.5±20.9) and dentin (Fig. 3c: 78.7±29.1). For the other materials, the performance on enamel and dentin was either dissimilar or antagonic. In the self-etch groups (Fig. 4a), no significant differences at the *total margin length* were detected between UniFil Bond (65.7±19.9), G Bond (78.8±11.9) and the exp. adhesive (69±15.6). Once again, a distinct behavior was observed with these materials when confronted to enamel (the case of the exp. adhesive with lower percentage of cont. margins, Fig. 4b, letters A,B) and dentin (the case of UniFil Bond with lower percentage of cont. margins, Fig. 4c, letter B). G Bond still presented the highest percentage of cont. margins on both enamel and dentin (60.9±21.5 and 98±2.9, respectively). Two-way ANOVA showed a significant interaction between the factors "tooth substrate" and "adhesive–composite" combination, indicating that the differences that existed between marginal adaptation in enamel and dentin were dependent of each material. In other words, some materials performed better on enamel, some on dentin and very few performed equally on both substrates. This was the case for iBond and G Bond; both are very similar in their composition and pH. Nevertheless, the highest results on marginal adaptation were observed on dentin for both materials (Fig. 4c) while iBond presented significantly lower percentage of cont. margins on enamel (Fig. 4b).

The average mean value attained by all groups on enamel and dentin at each testing interval is presented in Fig. 5. Both enamel and dentin margins degraded due to loading and further water storage. When compared to T0 (before loading), we observed an increased percentage (28%) of marginal gaps on dentin after thermomechanical loading and 1 year of water storage (T2). Enamel margins also suffered from degradation as an increased percentage (32%) of marginal gaps were observed after T2.

Discussion

This study evaluated the behavior of a large number of adhesive–composite combinations in a cervical restoration model with margins located on enamel and dentin. Twelve currently used adhesive systems and 12 resin composites recommended by the manufacturer to be used with each adhesive system were selected to be tested in this study. The decision to follow such protocol was not easy; while
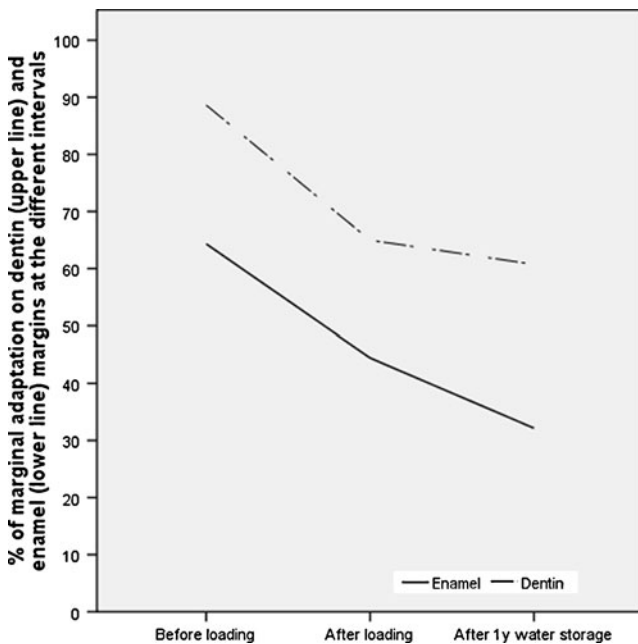
Fig. 5 Degradation of enamel and dentin margins across the three testing intervals. See that margins located on enamel and dentin suffered from degradation after each testing interval

some studies recommend to use an adhesive–composite combination from the same manufacturer [22–24], others showed that combining materials from the same manufacturer had no significant influence on bond strength [25, 26]. Unfortunately, these studies used bond strength as a testing method. Only one early report found high correlations between tensile bond strength, tensile strength, flexural strength and Young's modulus of eight commercial resin composites but found no correlation between these properties and marginal gap of restored cavities with the same composites [27]. The clinical relevance statement of this study was: "higher tensile bond strengths cannot be used to predict improved marginal adaptation of composite restorations" and this has also been confirmed in a recent study [28]. In addition, the main purpose of our investigation was not to compare the marginal performance of the different materials to search for the best one but to compare general properties, that is, the adhesion behavior on enamel and dentin, within the whole group. Therefore, we have followed manufacturer recommendations against mixing brands of composites and adhesive systems.

The rationale for using thymol as storage solution before cavity preparation was not arbitrary; it was our intention when preparing the protocol of this study, to follow the guidelines of tooth preservation in 0.1% tymol solution that were established several years ago by our research group [29–31]. Still, investigations on bond strength and microleakage in dentin indicated that thymol reduced adhesion to dentin [32–34] and that storage in chloramine T followed by storage for 2 h in distilled water prior to the experiment

[35] may be the first choice in tooth preserving solutions. Yet, the importance of storage solutions on enamel adhesion and on marginal adaptation is unknown. Studies that used the same methodology as the one in the present study, i.e., SEM quantitative margin analysis, have stored their specimens in varied liquids such as 20% ethanol [36], 0.5% chloramineT [37], 1% chloramine-B hydrate solution [38], 0.9% sodium chloride in water [39], 0.25% mixture of sodium azide in Ringer solution [40], 0.1 M thymol [41] and water [42]. In the context of our study, storage medium was not a variable because specimens from all groups were stored in the same solution. Therefore, comparison of our results with those of other authors that used different storage solutions should be carefully interpreted.

Class V cavities were selected as the experimental model because they are easy to perform. Additionally, the fact that enamel and dentin are present in the same cavity can provide with additional information on how a given material behaves when confronted to both substrates. Scanning electron microscopy (SEM) and quantitative margin assessment proved to be complementary evaluation methods. SEM analysis provided with microscopic details of the continuity of resin–enamel and resin–dentin interface. Margin analysis allowed the quantification of the rate of continuous or gap-free margins on both tooth interfaces. This technique has six interesting characteristics: 1. It is truly quantitative because numerical data is being collected by a trained technician who is observing the margins at 200× magnification, 2. The entire tooth/restoration interface is assessed, 3. It investigates both micromorphology and microleakage as 100% "excellent margin" (i.e., a perfect transition along the entire tooth/restoration interface), is associated with a perfect seal, 4. It is nondestructive as by analyzing gold-coated replicas, marginal qualities can be assessed both before and after exposure to thermomechanical stressing, 5. The method is highly discriminative, allowing the potential of different operative techniques to be quantified in terms of the percentage of "excellent margin" and 6. The technique is able to detect the early presence of an adhesive breakdown (marginal gaps) before catastrophic failures like loss of restoration's retention can occur. The presence of marginal gaps facilitates marginal coloration and microleakage. This is clinically relevant as both situations (marginal coloration and microleakage) usually require restoration replacement or retreatment on the mid-/long-term. Degradation resistance of the restoration margins was evaluated by the use of available methodologies with specific fatigue conditions and humid environment [43]. The specimens were subjected to thermomechanical loading and to additional 12 months of water storage. Thermomechanical loading provided us with the information on the materials' stress resistance under simulated oral conditions. It is well-known that materials

that are placed for long periods in the oral environment will undergo an interaction with oral fluids [44]. Therefore, additional water storage over a period of 1 year would give us information on the stability of these adhesives.

Fatigue process has been found responsible for the degradation of materials over time (Fig. 5). Restoration margins also suffer from degradation, the rate of degradation being material-dependent. This can explain the significant influence of thermomechanical loading and adhesive–composite combination on the marginal adaptation found in this study. Chemical changes or elution of resin components from the adhesive system due to water exposure might have been responsible for further increase of marginal gaps (Fig. 5). Cyclic chewing forces and thermocycling in the simulated oral environment induced fatigue at the adhesive interface, enabling microleakage and/or fluid permeation that probably accelerated the degradation process under water storage [45]. Although the factors "testing conditions" and "adhesive–composite combination" had a significant effect on marginal adaptation, the interaction between both factors was not significant. This means that none of the 12 combinations tested could be considered as superior over the others regarding the hydrolytic stability during a storage period of 12 months. Common degradation mechanisms might have affected the current materials tested [46].

A recent report found that resin–dentin interfaces surrounded by enamel resist better to hydrolytic degradation [13]. On the other hand, cavities with margins located in enamel and dentin are frequently found in clinical practice. The well-established concept that resin–enamel bonds are more durable than resin–dentin bonds [47] could not be corroborated in the present study. Increases of 32% and 28% marginal gaps (Fig. 5) were observed on enamel and dentin margins after one-year water storage, when compared with the initial results (T0). A previous study [48] also found up to 50% decrease in bond strength of resin–enamel interfaces after 4 h storage in water. Although a direct comparison between results are not suitable due to the use of different testing methodologies, this would support our findings that enamel–resin interfaces are also prone to chemical attack. Similar degradation mechanisms like the ones that occur on resin–dentin bonds are thought to be responsible for enamel bond degradation. Due to water sorption, delamination of the adhesive from enamel can occur due to water blisters present within the adhesive. Foxton et al. [49] described this phenomenon to explain the degradation on enamel–resin bonds observed in their study after 1 year of water storage, supporting our findings.

The observed *trend* that etch and rinse adhesives had higher percentages of continuous margins on enamel (Fig. 3b) and self-etching adhesives performed better on dentin (Fig. 4c) was demonstrated, for several, but not all

groups. This finding is in contrast with the common beliefs that etch and rinse adhesives usually perform better on enamel while self-etching adhesives perform better on dentin. Instead, we observed a material-related behavior that was independent of the adhesion strategy [50]. Said differently, the chemical composition of each material was one major determinant of the quality of adhesion to enamel and dentin and not the adhesion strategy, i.e., etch and rinse, self-etch, three-step, two-step, and one-step. Similar conclusions were recently reported in a 13-year clinical evaluation of class V restorations when etch and rinse and self-etching adhesives were used, supporting our findings [51]. In addition, a recent literature review [52] of 85 published class V clinical trials evaluated the annual failure rate, that is, the number of lost restorations per year, of class V fillings restored with 45 different adhesive systems belonging to all categories (three-step etch and rinse, two-step etch and rinse, two-step self-etch, and one-step self-etch). The authors found no statistically significant difference in the average annual failure rate between the different adhesive systems. However, within each category of adhesive, they did find variations in their performance that were explained by the composition of each material such as the presence/absence of filler, acetone, a certain type of copolymer, etc.

In the context of the present study, the materials' composition might have certainly accounted for the results. In the case of Optibond Fl, the highest results obtained on both enamel and dentin margins confirm those of other authors [52, 53] that rank this adhesive system as "gold standard". In the case of GBond, it is a HEMA-free adhesive and therefore, less hydrophilic and more hydrolytically stable if correctly applied. It is composed of the functional monomer 4-MET that has shown salt formation with hydroxyapatite [54]. It might explain why despite its high pH (around two), the highest results of marginal adaptation were obtained not only on dentin, but also on enamel. Seemingly, the well-known classification of adhesive systems (etch and rinse/self-etch, 3–2–1 step) is still useful for didactic purposes, but does no longer "speak" about the performance of a given adhesive, probably because manufacturers have been continuously improving the chemical formulations and new adhesive systems with better adhesion potential and simpler to use are no longer an ideal, but available materials. Our results confirm those of Blunck and Zaslansky [55] as they did not find, as well, any significant difference between G Bond and Optibond FL when enamel marginal adaptation was assessed on class I restorations.

Nevertheless, in addition to the chemical composition, results might be explained by a different behavior of the material when confronted to enamel and dentin within the same cavity. This behavior has, unfortunately, not been

detected in previous studies as most of the protocols dealing with adhesion use bond strength tests where enamel and dentin adhesion is usually tested in different teeth [56]. In the context of our study, among all the adhesive–composite combinations tested, only two groups (Optibond FL and G Bond) presented relatively equal percentages of continuous margins on enamel and dentin (Figs. 3 and 4). In the rest, different scores of marginal adaptation, within the same cavity, were observed on enamel and dentin. Said differently, if a higher percentage of continuous margins was observed on enamel, a lower percentage would be observed on dentin and vice versa. This proves that there is effectively a competition between enamel and dentin adhesion, especially when the restorative material is not able to efficiently bond to both substrates. In such a case, the restorative composite will shrink toward the superior bond at one margin and, at the same time, away from the weaker bond at the other margin [57]. These findings were recently corroborated by Chiang et al. [17] in their evaluation of the direction of polymerization vectors. They observed that shrinkage direction is affected by the adhesion of the filling material to enamel and dentin. When polymerization contraction stresses overcome the weakest attachment to the tooth substrate, the restoration surface is detached from this area and shrinks towards the area that offers the most durable adhesion. In the context of our study, this may explain why some adhesives performed better on enamel while others on dentin.

Interesting behaviors were the ones of iBond (chemical composition: UDMA, 4-MET, *glutaraldehyde*, acetone, water, photoinitiator, and stabilizer) and G Bond (chemical composition: UDMA, 4-MET, *phosphate monomer*, acetone, water, photoinitiator, and silica filler), whose chemical compositions are almost identical, except for *glutaraldehyde* that is contained in iBond and an additional *phosphate monomer* contained in G Bond. Both are HEMA-free and therefore more prone to phase separation, requiring careful drying of the solvent before polymerization [58]. Meanwhile, almost 100% continuous margins and no significant differences were detected between both materials on dentin, as shown in Fig. 4c. However, the results on enamel margins were significantly lower for iBond with respect to G Bond (Fig. 4b). Similar results were reported by Blunck and Zaslansky [55] in a recent evaluation of class I enamel margins. This was astonishing as both are one-step self-etching materials, have almost the same pH (G Bond 2.0 and iBond 1.8 [55], both are HEMA-free and very similar in their composition. Why one material performs better on enamel than the other is a good question. The authors speculate that it is due to the presence of glutaraldehyde and its effect on enamel adhesion. Söderholm et al. [59] tested different dental adhesives under shear bond strength and also found higher results on enamel with G Bond with

respect to iBond. These authors gave no explanation to their findings, as the main purpose of their study was to test the influence of six operators on the results. Blunck and Zaslansky [55] also observed significantly higher percentages of continuous margins on enamel with G Bond with respect to iBond. Malkoc et al. [60] also found a negative effect when a primer containing glutaraldehyde (Gluma Desensitizer) was applied over enamel. The authors argued that it was due to the presence of a layer of the antibacterial agent that contributed to incomplete resin penetration on the enamel surface. It is interesting to note that no information is available in the literature on this topic and it might be reasonable if we consider that when the first glutaraldehyde-containing adhesive was introduced by Asmussen in the mideighties [61], only multistep adhesives were available at that time and enamel etching with phosphoric acid was the only way to achieve mechanical interlocking on enamel. Today, with simplified self-etch adhesive systems, chemical components that in the past were applied only on dentin (like glutaraldehyde) are also applied on enamel. In addition, as enamel with self-etching material conditioning with phosphoric acid is no longer mandatory, these adhesive systems have to deal with a smear layer that is present also on enamel, as shown in a recent study [62]. It is impossible to prove, without more scientific evidence, if glutaraldeyde has a negative effect on enamel adhesion, but in any case this topic certainly needs further investigation.

Because 12 composite resins were used in this study, it is possible that differences in polymerization shrinkage of the materials tested may have influenced the results. Peutzfeldt and Asmussen [63] found, after testing 11 different composite resins, that viscous flow and polymerization shrinkage were significant determinants of *dentinal* gap formation. Takahashi et al. [64] corroborated these findings after testing eight different composite resins on *dentin* cavities and concluded that the adhesive system, polymerization shrinkage, viscoelastic properties and stiffness of the restorative material, cavity size and geometry, restorative placement and curing techniques were important determinants of marginal adaptation or gap formation. From the author's point of view, the important contribution of our study is that because our class V restorations had margins located on both enamel and dentin, we were able to demonstrate that for a given adhesive system and composite resin, marginal gap formation also depends, in addition to the aforementioned factors, on to which tooth substrate (enamel or dentin) the strongest and weakest adhesion will take place.

As mentioned above, the authors are certain that the chemical composition and mechanical properties of each material may have played an important role in the results [65, 66]. However, due to the design of the study where

different adhesive systems and restorative composites were tested, it is not possible to conclude whether results were due to the influence of a given adhesive system or restorative composite. Consequently, the present results apply for the material combinations tested in the present study. Therefore, the null hypothesis that there would be no difference in marginal integrity and degradation potential of enamel and dentin with the different materials tested must also be rejected. In materials with a similar performance at the total margin length, some will equally bond to enamel and dentin, and some materials will better interact either with enamel or with dentin.

In conclusion, thermomechanical loading and water storage had significant effects on marginal adaptation of all materials. Regarding hydrolytic stability that was indirectly assessed through marginal degradation, none of the materials could be considered superior than others after the water storage period, indicating that chemical degradation affects all restorative systems in a similar way. A distinct behavior was also observed in their bonding ability to enamel and dentin. This indicates that within the same restoration, only few materials can equally bond to both tooth substrates. From the clinical standpoint, the results showed that restoration margins will be more prone to degradation depending not only on the material that is used, but also on the nature of the substrate to which the material is bonded.

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