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ORIGINAL ARTICLE

Effect of different monomer-based composites and acid etching pre-treatment of enamel on the microleakage using self-etch adhesives systems

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Abstract

Objective. To evaluate quantitatively the marginal microleakage of restorations carried out with self-etching adhesives with or without prior phosphoric enamel acid etching of silorane or methacrylate resin-based composite restorations subjected to thermal cycling. ***Materials and methods.*** Forty cavities were prepared at the proximal surface of bovine incisors and randomly divided according to the etching of the enamel and restorative system used. The groups were restored with methacrylate [Adper SE Plus adhesive (3M ESPE) + Filtek Z250 (3M ESPE)] or silorane [Filtek LS adhesive (3M ESPE) + Filtek LS composite (3M ESPE)] restorative systems, light-cured using a LED unit (Bluephase 16i, Vivadent). After restorative procedure and thermocycling (1000 cycles), the specimens were immersed in methylene blue for 2 h. The specimens were triturated and the powder was used for analysis in an absorbance spectrophotometer. Data were statistically analyzed by 2-way ANOVA (alpha = 0.05). ***Results.*** No statistical difference between the restorative materials tested with or without previous acid etching of enamel in Class II marginal microleakage was observed ($p > 0.05$). ***Conclusions.*** The use of acid etching prior to self-etching adhesives did not interfere on the microleakage of methacrylate- or silorane-based restorations.

Key Words: *composite resins, dental leakage, dental restorations, methacrylate, silorane*

Introduction

The clinical use of composite resin as a restorative material has increased substantially in recent years because of excellent aesthetic properties and simplified bonding procedures [1,2]. However, the clinical longevity of restorations is dependent on the formation of effective bond interface between tooth and restorative material to control marginal microleakage [3,4]. The majority of dental composites utilized in clinical practice are based on the methacrylate chemistry [5]. The volumetric shrinkage resultant of the free radical polymerization in the methacrylate-based composites ranges from 2–5% [6]. Clinically, the composite strain is hindered by confinement of material bonded to the tooth and, as a result, shrinkage develops stress [7] around the tooth–restoration interface, increasing the likelihood of mechanical failure and permitting the ingress of bacteria, which may result in pulpal irritation or enamel microcrack propagation, enamel fracture and cuspal deflection [5].

Recently, a novel monomer system was developed, synthesized from the reaction of oxirane and siloxane molecules, termed silorane [5,6]. The ring-opening polymerization of this new composite instead of free radical polymerization of methacrylate monomers reveals low polymerization shrinkage [8], ~0.99 vol % [4]; moreover, it presents better biocompatibility, marginal adaptation and less microleakage than methacrylate-based systems [6].

The self-etch adhesive system is based on the absence of the steps of rinsing and drying, maintaining the ideal humidity of the dentine, thus its application is less technical sensitivity than total-etch systems [9]. However, additional selective enamel etching has been proposed when using this type of

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adhesive systems due to its lower demineralization compared with the phosphoric acid on this substrate [10], improving the marginal sealing quality [11–13].

The purpose of this *in vitro* study was to evaluate quantitatively the marginal microleakage of methacrylate and silorane monomer-based composites and the effect of the phosphoric acid etching of enamel margins using self-etch adhesive systems. Two null hypotheses were tested: (1) there would be no difference on the microleakage of methacrylate and silorane restorative systems and (2) the prior phosphoric acid etching of the enamel does not reduce the marginal microleakage.

Materials and methods

The information of the materials used in this study is collated in Table I. Forty sound freshly extracted bovine incisors were collected, cleaned with a periodontal curette, polished with a rubber cup and pumice paste under water and then they were stored in distilled water until they were used, within 1 month. The teeth had a part of their roots embedded in cold cure polystyrene resin (Piraglass, Piracicaba, SP, Brazil) and, afterwards, were split obliquely, 10 mm from the amelodentinal proximal junction using a double-faced diamond disc (KG Sorensen, Barueri, SP, Brazil). After cutting, they were finishing with water abrasive papers SiC 600-grit, to obtain a smooth and flat incisal surface.

Specimen preparation

Cavities were made using a diamond tip #3146 (KG Sorensen). All the cavities were prepared with a water-cooled high-speed turbine coupled to a unit of cavity preparation on the most plane proximal, simulating Class II, measuring 8 mm height, 4 mm of wide and 1.5 mm deep, under irrigation with air/water spray. The burs were substituted every five preparations. The cavities were randomly restored, following the manufacturer's instructions, according to the experimental groups. In the methacrylate and silorane groups with prior acid etching, the selective enamel etching was realized for 15 s using 35% phosphoric acid (3M ESPE, St Paul, MN), then washed for 15 s and dried thoroughly with mild airflow. For methacrylate groups the liquid A of the Adper SE Plus (3M ESPE) adhesive system was applied throughout the cavity, then two consecutive coats of liquid B were vigorously applied for 20 s, gently air-dried for 10 s after each layer of liquid B and light polymerization was carried out with a second-generation light-emitted diode (LED) unit for 10 s (Bluephase 16i; Vivadent, Bürs, Austria) at 1390 mW/cm^2 monitored by radiometer. For silorane groups, primarily the primer of Filtek LS System Adhesive (3M ESPE) was applied for 15 s, gently air-dried for 10 s and light cured for 10 s, then the bond was applied for 15 s, gently air-dried for 10 s and polymerized for 10 s. In the methacrylate groups, the microhybrid

Table I. Products, composition and application mode of the adhesives used in this study.

Products (Manufacturer)	Composition*	Application mode
Adper SE Plus Self-Etch Adhesive (3M ESPE, St Paul, MN)	*Liquid A*: Water, HEMA, Surfactant, Pink colourant *Liquid B*: UDMA, TEGDMA, TMPTMA, HEMA phosphates, Bonded zirconia nanofiller, Initiator system based on CQ	Apply Liquid A. Vigorously Apply Liquid B for 20 s. Dry with air stream for 20 s. Apply a second coat of adhesive. Dry with air stream for 20 s. Light cure for 10 s
LS Self-Etch Adhesive (3M ESPE, St Paul, MN)	*Primer*: BisGMA; HEMA; phosphoric acid-methacryloxy-hexylesters ethanol; water; silane treated silica; 1,6-hexanediol dimethacrylate; copolymer of acrylic and itaconic acid; (dimethylamino)ethyl methacrylate; phosphine oxide dl-camphorquinone *Bond*: substituted dimethacrylate; silane treated silica; TEGDMA; phosphoric acid-methacryloxy-hexylesters; CQ; 1,6-hexanediol dimethacrylate	Apply on tooth surface and brush primer for 15 s. Expose to a gentle air stream. Cure for 10 s Agitate the bond bottle. Apply on tooth surface. Expose to a gentle air stream. Cure for 10 s
Filtek Z250 composite (A2 shade; 3M ESPE, St. Paul, MN)	*Filler*: 60 vol%, aluminium oxide, silica, and zirconium oxide (0.01–3.5 μm) *Resin*: Bis-GMA, Bis-EMA and UDMA.	Insert in increments 2 mm thick each, approximately, and light-cure for 20 s
Filtek LS composite (A2 shade; 3M ESPE, St. Paul, MN)	*Filler*: 55 vol%, silica and yttrium trifluoride (0.04–1.7 μm) *Resin*: Bis-3,4-Epoxycyclohexylethyl-Phenyl-Methylsilane and 3,4-Epoxycyclohexylcyclopolymethylsiloxane	Insert in increments 2 mm thick each, approximately, and light-cure for 20 s

*As informed by manufacturers.
HEMA, 2-hydroxyethylmethacrylate; CQ, camphorquinone; Bis-GMA, bisphenol-A glycidyl dimethacrylate; TEGDMA, triethylene glycol dimethacrylate; Bis-EMA, bisphenol-A ethoxylated dimethacrylate; UDMA, urethane dimethacrylate.

composite resin Filtek Z250 (3M ESPE) and in the silorane groups, the microhybrid composite Filtek LS (3M ESPE) was inserted in four horizontal increments 2 mm thick each, approximately, and polymerized for 20 s. After 24 h of storage in water at 37°C, the restorations were finished and polished with medium, fine, and superfine aluminum oxide discs Sof-Lex Pop-on (3M ESPE), in decreasing order of granulation.

Thermal cycling

The groups were submitted to ageing test, thermal cycled 1000 times (5 ± 2°C and 55 ± 2°C) with a dwell time of 1 min at each temperature and a transfer interval of 5 s.

Dye immersion

After these procedures, the entire specimens (except the tooth–restoration interface) were protected with two layers of fast setting cyanoacrylate-based adhesive Superbonder (Henkel Loctite Adhesives LTDA, Itapevi, SP, Brazil). Before dye immersion, a 1 mm strip of adhesive tape was placed around the area that was infiltrated, and two layers of nail varnish were applied. Then, the specimens were totally immersed in 2% neutral methylene blue solution (Proderma, Piracicaba, SP, Brazil) for 2 h. After this period, the blocks were removed from dye solution, washed in running water and dried. The nail varnish was removed with the use of a periodontal curette and the dye on the restoration was removed wearing off 0.05 mm from the surface, controlled by a caliper.

Sample trituration

To take a reading of the infiltrated dye colour, specimens (dental block + restoration) were initially split up and weighed. After the weighing the specimens were triturated in a hard tissue mill (Marconi Equip. Ltda., Piracicaba, SP, Brazil) in order to obtain a powder composed of tooth/restoration and then weighed again. If the difference between the initial and final weight were higher than 10%, the specimen would be discarded. In this study, no specimen was discarded.

Dissolution

After trituration, the powder obtained from each sample was separately immersed, in test tube, containing 4 ml of absolute alcohol for analysis (Merck, Darmstadt, Germany), for 24 h, to dissolve the dye that leaked through the tooth/restoration interface. The solution obtained was centrifuged at 3000 rpm for 3 min (Tomy IC 15AN, Tomy Ind., Tokyo, Japan), so that the powder and other elements decanted. The supernatant of the centrifuged solution was submitted to quantitative analysis of the dye present in the solution by a spectrophotometry (DU 65, Beckman–Instruments, Inc., Fullerton, CA) unit through absorbance reading.

The absorbance reading was taken in an adjusted unit at a wavelength of 668 nm, corresponding to the maximum absorbance of methylene blue dye. Prior to the readings, the spectrophotometry unit had been adjusted by spectral reading with pattern solutions at the concentrations of 0; 0.1; 0.2; 0.3; 0.5; 1; 2; 4 μg/ml, to obtain the maximum spectral absorbance wavelength. Readings of the solutions were made using the wavelength value to find the maximum value of spectral absorbance. Through the ABS-concentration system, one obtains the r^2 value (0.9996) and the equation of the line ($y = a + bx$). The following regression was obtained: Absorbance = 0.2716 × (dye concentration) − 0.0075. From this regression, dye concentration could be calculated. A graph of lines in a Cartesian system of axes was drawn, using the values of dye concentration in micrograms per millilitres on the axis of the abscissas and the optical density obtained on the axes of the ordinates. The linear regression was obtained from Y as a function of X to determine the equation of the line, from which the concentration of dye was calculated.

The microleakage data of experimental groups was submitted to two-way analysis of variance ANOVA and Tukey's test. The level of statistical significance was pre-set at 5%.

Results

The results showed no difference on the marginal microleakage between restorative systems tested ($p > 0.05$) and the use or not of previous phosphoric acid etching of enamel margins ($p > 0.05$) in Class II restorations (Table II), as well as between the factors interaction ($p > 0.05$).

Discussion

The application of self-etch adhesive systems with or without prior enamel etching for methacrylate and silorane resin-based restorations showed no difference on the microleakage. Therefore, the two null hypotheses were accepted. The low-shrink composite material based on silorane does not behave differently from methacrylate composite material, besides the lower polymerization shrinkage of the silorane. This result disagrees with studies that showed that the microleakage of silorane is lower than that of methacrylate-based composite [6,12]. A possible explanation is the use of an incremental restorative technique, an approach used to minimize the effects of curing shrinkage [6].

Table II. Microleakage (μg/ml) means (standard deviation) according to restorative system and previous acid etching of the enamel margins before adhesive procedures.

	Enamel acid etching	
Restorative system	Without	With
Methacrylate	0.0357 (0.0122)	0.0357 (0.0149)
Silorane	0.0394 (0.0138)	0.0362 (0.0058)

There is no statistically difference between the restorative systems ($p > 0.05$) and with or without prior enamel acid etching ($p > 0.05$).

The use of incremental insertion of composite has been studied [6,14,15] and despite there being no consensus among the different insertion techniques (obliqual, horizontal, vertical) there is an agreement about the positive effect on polymerization composite stress relief. The decrease in shrinkage stress is due to minimal contact with the cavity walls during polymerization as well as the reduction of the shrinkage produced by a small volume material, an affirmation valid for each individual increment [15]. It could be inferred that the incremental technique used to restore the Class II cavity was able to minimize the effect of confinement on the contraction stress development and the challenge on the bond interface, but the filling technique used the smallest number of composite increments possible, once the sum of residual shrinkage of the increments can increase the cuspal strains, and increase the amount of stress accumulation [16,17]. However, in this study four increments of 2 mm of thickness each was used to obtain an adequate polymerization rate.

The effective sealing of the cavities walls also depends upon the performance of the adhesive layer. Depending of the etching aggressiveness, self-etching adhesives can be sub-divided into strong (pH <1), intermediary strong (pH ≈ 1.5), mild (pH ≈2) and ultra-mild (pH >2.5) showing different interaction patterns at the enamel and dentin compared to phosphoric acid treatment after the etch-and-rinse approach and also with each other [18,19].

In the present investigation, two restorative systems were evaluated. The methacrylate-based composite was used with a 'strong' (pH <1) acid two-bottle self-etch bonding agent, consisting of an aqueous primer and an acidic adhesive. According to the manufacturer, the separation of the aqueous primer and acidic components minimizes hydrolysis of the acidic phosphates for improved shelf stability. Strong self-etch adhesives present deep demineralization effects at both enamel and dentin and the dissolved calcium phosphates are not rinsed away. These embedded calcium phosphates are expected to be very unstable in an aqueous environment and a serious weakening of the interfacial integrity could be expected [19].

The silorane-based composite was used with the proper two-step 'ultra-mild' self-etch bonding agent (pH ≈2.7). The first step, self-etching primer is applied and light-cured. This self-etch primer contains phosphorylated methacrylates, BisGMA, HEMA and water/ethanol as the solvent. Then, the bond agent is applied and light-cured for 10 s. Some studies reported that mild self-adhesives in non-instrumented enamel might be unsatisfactory [12,20]. Indeed, clinical research has revealed that marginal defects at the enamel margins of a composite restoration develop rather rapidly, whereas the dentin margins appear to maintain their marginal integrity much longer [11].

It has been suggested that the selective enamel etching can improve the bond strength [21,22] and decrease gap formation and microleakage [12] of self-etch adhesives. However, the results of the present study do not support this hypothesis since the use of prior etching with phosphoric acid did not improve the sealing and there was no difference in microleakage, irrespective of the restorative system used. The methacrylate restorative system uses a strong self-etching adhesive (pH <1), which seems to promote a demineralization pattern that resembles the one obtained when the enamel is etched with phosphoric acid [19]. For silorane restorative system, although an ultra-mild self-etch bonding agent (pH ≈2.7) was used, there was no difference with or without prior etching as well, whilst this one could be expected. This result could be due to the hydrophobic resin coat applied, which might improve the bond durability acting as a stress absorber layer. The hydrophobic structural network polymer probably maintains a favourable sealing behaviour after fatigue stress [13,20,23].

For the four experimental groups, no difference was observed in microleakage and it seems that all advantages and shortcomings of each self-etching system used with or without prior treatment with phosphoric acid lead to a situation with gains and losses and we conclude that there is no definite protocol to be dictated. In an *in vitro* study, Reis et al. [24] demonstrated that the increased application time and/or association of the etch-and-rinse and self-etching techniques did not improve bond strength for ground enamel. Indeed, we agree with the previous authors [24] who stated that the additional step to pre-treat the enamel surface contradicts the intent of the simplified approach of self-etching systems and increases the complexity of the procedure without bringing improvements in the marginal sealing.

Conclusions

Within the limitations of the current study, the following conclusions were drawn: (1) none of the restorative systems tested totally prevented marginal

microleakage and (2) additional phosphoric acid etching of the enamel margins did not reduce the microleakage.

Declaration of interest: The authors report no conflicts of interest. The authors alone are responsible for the content and writing of the paper.

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