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ORIGINAL ARTICLE

Marginal adaptation of three self-adhesive resin cements vs. a well-tried adhesive luting agent

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Abstract This in vitro study compared the marginal adaptation of three self-adhesive composite cements with the clinically well-tried dentin adhesive system Panavia F 2.0. A total of 32 Empress 2 all-ceramic MOD-inlays (eight in each group) were luted using the self-adhesive composite cements Maxcem, Multilink Sprint, and RelyX Unicem Clicker; Panavia F 2.0 served as a clinically well-tried control. Each luted inlay underwent long-term water storage of 90 days as well as additional mechanical and thermal loading to simulate oral service. Marginal integrity was evaluated in both dentin and enamel finishing lines using scanning electron microscopy (SEM) and dye penetration tests. Dye penetration was lowest for Panavia followed by RelyX Unicem. Maxcem and Multilink showed a considerable dye penetration of up to 60%. After aging, SEM analysis revealed a reduction of "perfect margin" areas for Multilink Sprint and RelyX Unicem in enamel and for Maxcem and Multilink in dentin. Compared with the well-tried system Panavia—which was assumed as the golden standard of adhesive luting systems—only the self-adhesive luting agent RelyX Unicem showed similar results of marginal adaptation after long-term water storage.

Keywords Self-adhesive · Luting agents · Aging · Adhesion · All-ceramic inlays · Long-term water storage

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Introduction

A resin-based luting agent simply bonds to enamel by etching the hydroxyle-apatite, whereas the bond between dentin and resin cement results from a complex procedure [1–3]. Whenever dentin is processed, its surface will be covered by a smear layer that consists of bacteria and an organic or inorganic excess of dentin. The smear layer can be removed by etching only. Nakabayashi described a method for etching dentin followed by the infiltration of hydrophilic monomers into the exposed collagen network and opened dentin tubuli [3]. The infiltrated monomers were coated with a hydrophobic resin and polymerized, forming a micromechanical interlocking called "hybrid layer," to which layer composite cement can be applied for bonding the restoration.

Meanwhile, different dentin adhesive systems are available, which may be classified as tooth-conditioning systems:

1. with simultaneous enamel/dentin phosphoric acid etching and rinsing prior to the application of multibottle or one-bottle adhesives
2. with selective phosphoric acid etching and rinsing on enamel and selective self-etching systems on dentin
3. containing acid monomers, which may simultaneously condition both enamel and dentin without rinsing [2]

Dentists may be perplexed by this variety of products and their different conditioning steps. To simplify tooth-conditioning procedures, all-in-one adhesive systems without mixing and rinsing have been developed. However, literature reports demonstrate that multibottle systems with simultaneous etching and rinsing show superior in vitro and in vivo activities compared to the new all-in-one systems [4–6]. Blunck stated in his study on class V cavities that "the results from the enamel margin show that the all-in-one adhesives seem to be significantly affected by water storage" [4]. Kraemer and Frankenberger found in an in-vitro study on class II cavities that "etch and rinse adhesives combined with conventional luting resin composites reveal still the best prognosis for adhesive luting of glass ceramic inlays" [5]. These in vitro data are supported by clinical results. Restorations with extensive dentin-bonded ceramic coverage have also been successful [6]. Of the nonrinsing systems, Panavia has been proven in in vivo [7, 8] and in vitro studies [9, 10] to reliably bond to enamel and dentin.

Recently, the concept of a self-adhesive cement has been launched into the market [9, 10]. Dentists only have to clean and dry the tooth surface. This type of cement should simultaneously etch enamel and dentin and be initiated by light or autocuring systems. The first material used was RelyX Unicem (3M ESPE, Seefeld, Germany), which showed promising first in vitro results [9, 10]. In vivo data are still rare [11, 12]. Meanwhile, more manufacturers offer self-adhesive cements. This study aims at comparing marginal adaptation on enamel/dentin-to-cement and cement-to-restoration interfaces of these new cements after 90 days of water storage [4] followed by additional mechanical and thermal aging. Marginal adaptation is characterized by semiquantitative scanning electron microscopy analysis and dye penetration test. Results are compared to those of a well-tried standard adhesive system. The hypothesis was that the marginal adaptation of self-adhesive luting agents and a standard adhesive system did not differ after aging.

Materials and methods

Thirty-two human extracted molars were used, stored in a 0.5% chloramine solvent for no more than 6 weeks. Teeth roots were coated with a layer of polyether measuring approximately 1 mm in thickness (Impregum, 3M ESPE, Seefeld) and axially inserted into poly(methyl methacrylate) (PMMA) resin (Palapress Vario; Heraeus-Kulzer, Wehrheim, Germany) [13]. The polyether layer simulated the physiological tooth movement capacity given by the periodontium. The inserted teeth were randomly assigned in four subgroups of eight teeth each. A class II (MOD) inlay cavity was prepared using diamond burs. The finishing lines of proximal boxes were mesially in dentin and distally in enamel. Impressions were taken from the prepared teeth (Permadyne-Penta, 3M ESPE, Seefeld), and gypsum casts were made. On the casts, Empress 2 all-ceramic inlays were manufactured by a dental technician of the department. The three-point occlusal contact area was constructed to correspond to the human molar antagonist. The antagonists were later used to apply the occlusal load during simulation of oral stress.

Prior to the setting, the inner surface of the crowns were etched for 20 s with hydrofluoric acid (IPS Ceramic etching gel, Ivoclar-Vivadent, Schaan, Liechtenstein), and a silane coupling agent was applied (Monobond S, Ivoclar-Vivadent). The cementation process and details of the luting agents used are depicted in Table 1. All cements were light-cured for 40° (each side) using the Elipar light-curing device standard mode (3M ESPE, Seefeld).

The inlays were finished and polished with Sof-Lex discs (3M ESPE, St Paul, USA). Impressions of the entire marginal area were taken with a polyether material (Permadyne-Penta, 3M ESPE, Seefeld), and epoxy-resin replicas were cast (Epoxy Die, VP 1031, Ivoclar-Vivadent) for scanning electron microscope (SEM) marginal analysis. All inlays were first stored in distilled water at 37°C for 90 days, followed by thermal cycling and mechanical loading (TCML): 6,000×5/55°C, changing every two minutes, $1.2\times10^{6}\times50$ N, 1.66 Hz. These parameters should represent a 5-year period of oral stress [14–16]. The entire simulation process lasted 8.3 days. Thus, the samples stayed in water for 90 days [4] and in the masticator for 8.3 days. The load of 50 N was applied by the same third human antagonists, which were used to form the occlusal contact area of the

Table 1 Cementation process and details of the luting agents

Cement	Batch #	Pretreatment of tooth surface	Etching system
Maxcem (Kerr Hawe, Orange, CA, USA)	# 420326	No	Glyceroldimethacrylate dihydrogen phosphate
Multilink Sprint (Ivoclar-Vivadent, Schaan, Liechtenstein)	# J 26111	No	Phosphone acid
Panavia F 2.0 + ED primer (Kuraray, Osaka, Japan)	# 41214	Mixing A/B Primer (1:1)	ED primer I: MDP, hydrophilic phosphate group
		Primer application for 20 s	ED primer II: Hydrophobic alkyl group double bond end, phosphate monoester, campherquinone
RelyX Unicem Clicker (3M ESPE, Seefeld, Germany)	# 274070	No	Phosphoric-acidic methacrylate

inlays. Thus, the load was assured to be axially inserted with a three-point support. After water storage and TCML, impressions were taken again from the marginal areas to construct epoxy resin replicas.

Semiquantitative investigation of marginal adaptation

The analysis of SEM-images was conducted with the Optimas 6.2 image analysis system (Optimas, Orange, USA).

Restoration-to-cement and cement-to-enamel/dentin interfaces were examined. Assessment criteria were defined as follows [17, 18]:

1. Perfect margin: The two adjoining surfaces show no interruption of the continuous margin and merge into each other without any difference in level.
2. Marginal gap: The two adjoining surfaces show slight imperfections with interruptions in continuity, the forming of gaps or cracks due to loss of cohesion or adhesion.
3. Non-assessable areas: All adjoining areas that do not fit criteria 1 and 2.

Dye-penetration test

After TCML, the polyether layer was removed from all teeth roots [19]. With the exception of the proximal margin area, the teeth were completely coated with nail varnish to prevent dye penetration elsewhere. The coated teeth were stored in 0.5% basic fuchsine solution at 37°C for 16 h. After rinsing off the fuchsine solution with distilled water, the teeth were embedded in PMMA resin (Palapress Vario, Kulzer), luted on a tray of a low-speed diamond saw (Leica SP1600, Leica, Nussloch, Germany), and cut into 500 μm slices (eight slices per tooth) in an oro-vestibular direction. The slices were mounted on transparent foils. Percentage of dye penetration was measured from both sides using a microscope (×25; Zeiss, Oberkochen, Germany) and the image analysis system Optimas 6.2 (Optimas).

Statistics

Means and 95% confidence intervals of means were calculated. Statistical differences were established using analysis of variance and Tukey tests. The level of significance was set at α=0.05.

Results

Scanning electron microscopy of enamel

After aging, Multilink Sprint and RelyX Unicem showed a lower number of perfect margin areas in comparison to Maxcem and Panavia F 2.0. The difference was not statistically significant (post hoc $p<0.05$). Values were widely distributed (see Fig. 1a), and the 95% confidence interval had the lowest values of 80% of perfect margin integrity for Multilink and RelyX Unicem. No investigated luting agent demonstrated any different behavior on inlay–cement or cement–enamel interfaces (Fig. 1a).

Scanning electron microscopy of dentin

In dentin, the lowest percentage of perfect marginal adaptation after aging was found in Maxcem and Multilink Sprint. The deteriorations were not statically significant (see Fig. 1b), but both cements showed a wide distribution

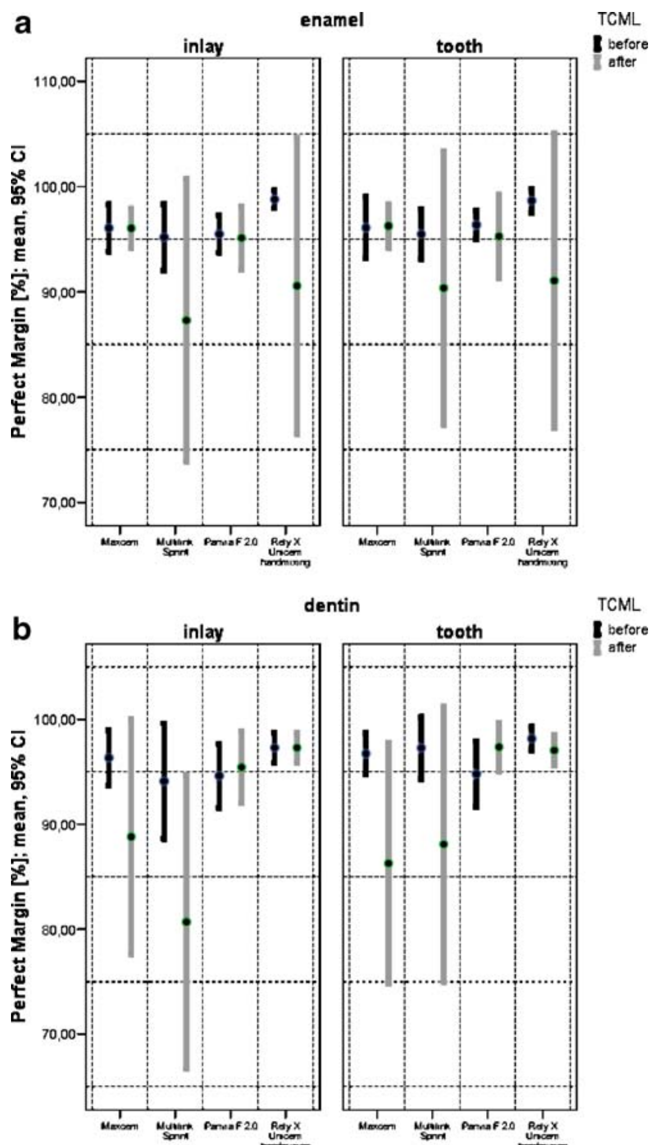

Fig. 1 **a**, **b** Semiquantitative analysis of the criteria perfect margin of finishing lines in enamel and dentin

of values. The 95% confidence interval of means showed the lowest values of up to 70% of perfect margin areas for Multilink on inlay–cement interfaces. No investigated luting agent bonded differently to inlay–cement or cement–dentin interfaces.

Dye penetration

According to Fig. 2, luting agents can be ranked as follows: the lowest dye penetration showed in both enamel and dentin for Panavia F 2.0, followed by RelyX Unicem, Maxcem, and Multilink Sprint. In enamel, Multilink Sprint and Maxcem showed a significantly higher dye penetration than RelyX Unicem and Panavia ($p<0.05$). The means of Multilink reached values of 60%, and the 95% confidence interval went up to more than 75% of dye penetration after aging.

In dentin, only Maxcem showed statistically significant higher penetration values than Panavia. For Maxcem, the mean dye penetration was higher than 50%, and the confidence interval showed that the highest values may be 65% and more.

Discussion

This study uses a high-resolution method for the quantitative analysis of the marginal adaptation of luting agents. Dye penetration and semiquantitative marginal analysis using human extracted teeth have their limitations, as described in the literature [17–19]. The advantage of using human teeth is that adhesive cementation may be studied. However, variations between the structure of hydroxylapatite and the dimension and the history of the teeth result in higher standard deviations. But when the same operator evaluates the same specimens twice, the difference between results is less than 4% [17]. For this reason, the same operator evaluated the samples before and after the aging procedure, and the chosen method can be used to evaluate marginal integrity.

Marginal adaptation is influenced by different parameters, such as shrinkage of the luting agent, the c-factor, insertion technique, and polymerization protocol [20–22]. High-resolution marginal analysis has been reported to provide indicators for the ability of adhesive systems to compensate shrinkage of resins during polymerization [21]. To introduce precise and repeatable stress, 6,000 thermal cycles with 5°C/55°C were conducted, which lasted 8.3 days. Therefore, an investigation using only this short period of water storage may fail to indicate effects of long-term water storage. Blunck and coworkers [4] emphasized that, in enamel, one-bottle all-in-one adhesives seemed to be significantly affected by water storage. There are reasons to assume that the new self-adhesive luting agents work with similar bonding concepts as the one-bottle all-in-one adhesives. To demonstrate possible effects of water storage, this investigation combined 90 days of water storage in distilled water at 37°C with thermal and mechanical loading of 8.3 days.

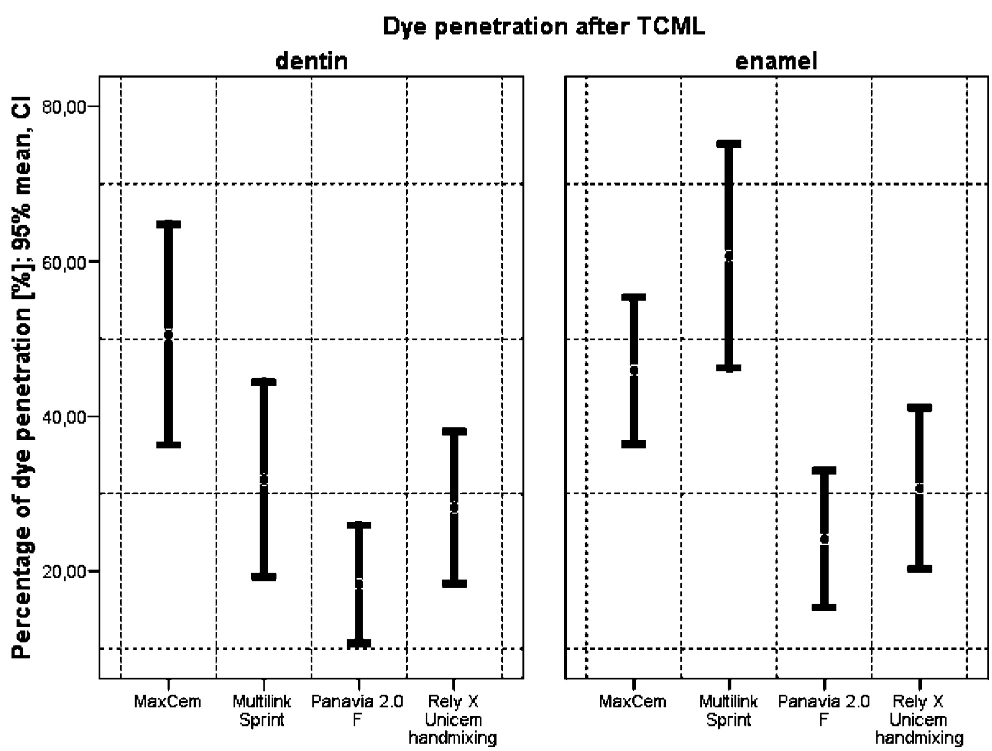

Fig. 2 Dye penetration analysis of areas with finishing lines in dentin or in enamel

The good clinical performance of Panavia for luting restorations is described in the literature [7, 8, 23]. Both SEM-analysis and dye penetration tests reflect the clinical behavior of this luting agent. This cement may therefore be used to rank other luting agents. From the three self-adhesive-cements evaluated, RelyX Unicem seemed to have a similar performance as Panavia. Meanwhile, the first 3-year clinical data of RelyX Unicem are available, confirming the results of this investigation. The only weak point of RelyX Unicem may be its adhesion to enamel. SEM analysis and dye penetration tests revealed the worst results of marginal adaptation for RelyX Unicem in enamel. This result agrees with literature reports describing RelyX Unicem to work better on dentin than on enamel surfaces [24, 25].

In contrast, Maxcem seems to perform better on enamel surfaces. After aging, the means of the criterion "perfect margin" were lower, and dye penetration was higher in dentin. However, the differences were not statistically significant.

Multilink Sprint showed the worst performance of all evaluated cements on all surfaces and interfaces. Mazzitelli assumed that the acidic monomers of Multilink Sprint were not properly neutralized so that they retain their etching potential, affecting the polymerizing reaction and jeopardizing adhesion in long-term water storage [26]. Another explanation may be that its phosphone acid base etching system absorbs water and therefore jeopardizes adhesion.

Conclusion

The new self-adhesive luting agents cannot be considered as a consistent group of cements. Marginal adaptation in dentin or enamel after long-term water storage and thermal and mechanical loading of two self-adhesive cements do not reach the levels of well-tried multi-step adhesive systems. Only one self-adhesive resin cement showed results comparable to the golden standard system used in this study.

Conflict of interest statement The authors declare that they have no conflict of interest.

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