Silorane low shrinkage composite

Evaluation of selective features

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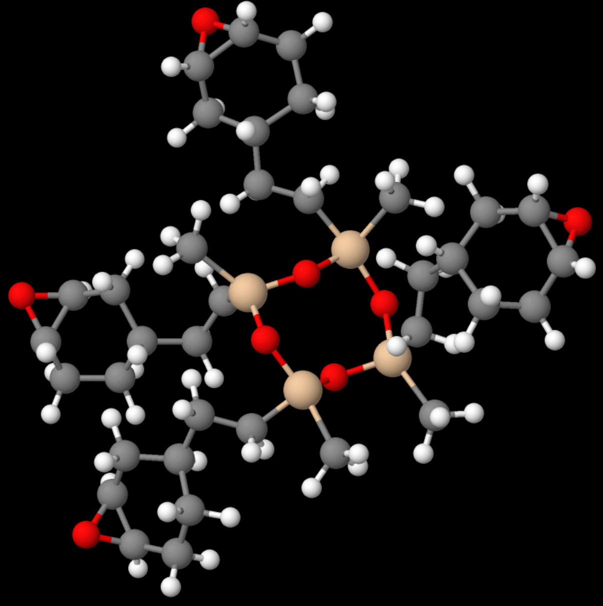
evaluation of selective features



Ladislav Gregor

2,4,6,8-tetramethyl-2,4,6,8-tetrakis[2-(7-oxabicyclo[4.1.0]heptan-4-yl)ethyl]-1,3,5,7,2,4,6,8-tetraoxatetrasilocane

Silorane low shrinkage composite: evaluation of selective features

Ladislav Gregor

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Silorane low shrinkage composite: evaluation of selective features

ACADEMISCH PROEFSCHRIFT

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prof. dr. ir. K.I.J. Maex
ten overstaan van een door het College voor Promoties
ingestelde commissie,
in het openbaar te verdedigen in de Agnietenkapel
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Ladislav Gregor
Geboren te Tábor,Tsjechië

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Faculteit der Tandheelkunde

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CONTENTS

Chapter 1

General introduction

1.1 Introduction

Restorative dentistry has undergone an important paradigm shift during last two decades. Due to increased emphasis on preserving healthy tooth tissue the early concept of amalgam and cast restorations based on macro-mechanical retention coupled with inevitable tissue lost was replaced by the philosophy of minimally invasive dentistry. The introduction of predictable adhesive technologies has made this concept achievable. An adhesive tooth restorative technique enables diseased or lost tooth tissue be replaced by adhering the restorative material directly to the remaining sound tooth tissue. Due to their improved esthetic qualities, strength and wear resistance, resin composites are nowadays the restorative materials of first choice for replacement of lost natural tissues.

It is generally accepted, that polymerization shrinkage of resin composite is a still unsolved problem in clinical dentistry [1]. Therefore, the main objective of this project was to evaluate selective properties of a new low shrinking composite and to compare them with commonly used ones.

1.2 Resin composite

Dental resin composites are versatile materials whose usage has continued to grow since their introduction to the profession over the last 50 years. They are used for a variety of applications in dentistry, including but not limited to restorative materials, cavity liners, pit and fissure sealants, cores and buildups, inlays, onlays, crowns, provisional restorations, cements for single or multiple tooth prostheses and orthodontic devices, endodontic sealers, and root canal posts. The composition of these materials is comparable as they are all composed of a polymeric matrix, reinforcing fillers, typically made from radiopaque glass, a silane coupling agent for binding the filler to the matrix, and chemicals that promote or modulate the polymerization reaction. Dental resin composites can be distinguished by differences in formulation.

There are four matrices on the market today: compomer-based, methacrylate-based, ormocer-based and silorane-based. Compomers consist of two main components: dimethacrylate monomer(s) with carboxylic groups and filler that is similar to the ion-leachable glass present in glassionomer cements [2]. Methacrylate-based resins are the most commonly used matrix materials in composites. A modification of this matrix is represented by ormocers, where the methacrylate-based resin is modified by the addition of small polysiloxane particles. A completely different chemistry is represented by the silorane matrix. This matrix is based on molecules consisting of siloxanes and oxiranes, therefore called ‘siloranes’, with a very hydrophobic characteristic. During the polymerization the matrix represented by the monomers transforms to a polymeric network. This process might be light-activated, by a photosensitive polymerization initiator or in a dual cure formulation containing two pastes (base and catalyst paste) that are mixed to start the chemically cure (CC), while exposure to light also can add to the polymerization reaction by activating the photosensitive initiator. The most common photo-initiator system for the methacrylate-based composites is camphoroquinone, accelerated by a tertiary amine, typically an aromatic one [3].

The second fundamental component in resin-based restorative materials is represented by fillers. Fillers can be divided depending on their size as macro fillers ($X > 0.4$ µm) and micro fillers ($X < 0.4$ µm). In case the fillers particles are fabricated by means of nano-technology, the corresponding resin composite may be denominated as nano-modified. When the microfiller particles are smaller than the wavelength of visible light, thus being invisible to the human eye, they give to the restorative materials a high and durable surface gloss [4]. Micro-filled resin composites have a low filler load, which is reflected in a relatively low Young’s modulus and fracture strength. As a consequence they are prone to chipping and fracture [5]. For this reason they are proposed mainly as veneering material in anterior restorations yet. Macro-filled composites are in general highly filled resins, characterized by high stiffness, hardness and compressive strength. Unfortunately, large fillers tend to increase wear of the material and exposure of filler particles because of resin matrix wear results in a higher surface roughness limiting the use of macro-filled composites as a base under other restorations or as a core under prosthetic restorations.

A good compromise between the high mechanical properties of macro filled materials and the good aesthetic properties of micro filled materials can be found in hybrid materials. They couple the necessity of being resistant to masticatory forces with the aesthetic requirements of modern dentistry. These characteristics confer to this family of materials a large indication both in anterior and posterior areas. That is why they are currently most commonly used and produced ‘multi-purpose’ restorative materials.

1.3 Adhesion

Adhesive dentistry is based on the bonding to various substrates. When making a direct restoration, the tooth colored resin composite might be bonded to two different substrates i.e. enamel and dentin. Performed since the 1950s, adhesion to enamel became immediately successful through micro mechanical retention created by acid etching and the application of a low viscosity hydrophobic resin [6]. Adhesion to dentin is rather complicated due to its composition, an organic matrix (collagen fibers), a mineral phase (calcium-phosphates) and water. The effectiveness of dentin adhesion depends on two critical steps, which are demineralization (etching) and application of hydrophilic monomers to the remaining organic matrix (bonding), which allows the development of an inter-diffusion zone (hybrid layer) to be created [7].

Based upon the underlying adhesion strategy, two types of adhesive systems are in use nowadays. When perform the etch&rinse approach both enamel and dentin are decalcified by using 30 - 40% H_3PO_4. The smear layer dissolves by the etching agent which is subsequently rinsed out by a water spray. Removing the smear layer and opening the underlying dentin tubules expose the delicate collagen fibers that are subsequently infiltrated with a primer, which enhance the wettability for the application of a resin adhesive. As such a ‘hybrid’ layer is formed that is composed of a mix of adhesive resin, collagen fibers and penetrated porous dentin. The adhesive strength is therefore determined by the strength of the hybrid layer. Simplified adhesive systems consist still of an etch&rinse phase followed by an application of a combined primer and adhesive liquid. Rather than removing the smear layer, the self-etch approach involves the application of an non-rinse acidic primer that will promote micro-mechanical bonding of the adhesive to the partly dissolved smear layer and the collagen of the underlying dentin [8]. This technique minimizes the potential for postoperative sensitivity by preventing the collapse of the collagen fibers that can occur after conditioning and drying in the etch&rinse process. Similar to etch&rinse adhesives, simplified adhesives that combine the (self-etch) primer with the adhesive resin were developed, one-step self-etch adhesives or so-called “all-in-one” adhesives. Etch-and-rinse adhesive systems, generally perform better on enamel than self-etching systems which may be more suitable for bonding to dentine [9].

1.4 Polymerization shrinkage

During free-radical polymerization the conversion of methacrylate monomer molecules into a polymer network results in a closer packing of the molecules leading to volumetric shrinkage [10]. This polymerization shrinkage creates contraction stresses that builds up at the interface of the restorative material and cavity walls [11]. Polymerization shrinkage stress may cause deformation of the tooth [12], open pre-existing enamel microcracks [13], or even initiate microcracking of the restorative material [14]. On the composite-tooth interface polymerization shrinkage stress can initiate adhesive failures which may cause microleakage, marginal discoloration, post-operative sensitivity or even the loss of the restoration [15].

The magnitude of the stress depends on a number of factors including volumetric shrinkage [16], the modulus of elasticity of the composite [17], it's coefficient of thermal expansion, bonding of the filler particles to the resin and their nature [18], curing characteristics [19], configuration of the cavity into which the restoration is placed (C-factor) [20] and compliance of the remaining tooth structure.

A number of clinical techniques comprising various incremental layering techniques [21-23], application of low-modulus intermediate layers [24], use of the different light curing protocols [25] have been described in the literature to help the clinician in reducing the effects of the inherent polymerization shrinkage stress. However, these strategies are often difficult to execute, time consuming and laborious.

1.5 Low shrinkage composite

As the polymerization shrinkage and related shrinkage stress development are considered to be major drawback in resin bonded resin composite restorations (RBCs) [1]. the research focuses on the development of low or even non (zero) shrinking materials, which would allow for simpler, faster and more reliable restorative techniques. The volumetric shrinkage of commercially available methacrylate based composites varies from 4.0 – 5.5vol.% (flowable composites 45-67 wt% filler loaded.) to 1.9 – 3.5 vol% (hybrid composites 74-79 wt% filler loaded). Several modifications of resin composite composition have been proposed for the reduction of volumetric shrinkage and contraction stress development. One possibility to reduce shrinkage of composites is the addition of different types of pre-polymerized particles like nano-element aggregates or pre-polymerized splinters. The second possibility is the increase of the filler load. Very highly filled systems like packable posterior composites or materials with optimized filler load up to 82 wt% reveal shrinkage values down to 1.7 vol% [1]. Unfortunately both factors have limitations, coming among others from handling properties [26] and elastic modulus [27].

Current changes are more focused on the polymeric matrix of the material, principally to develop systems with reduced polymerization shrinkage, and perhaps more importantly, reduced polymerization shrinkage stress formation. Polymerization shrinkage of methacrylate based monomers is highly influenced by the molecular weight of the monomer. Increasing molecular weight of the monomer (the larger the molecule) may reduce shrinkage. Recently several new resin composites based on high molecular weight monomers have been introduced to the dental market. The modified urethane dimethacrylate resin DX511 (Dupont) found in Kalore (GC), the urethane monomer TCD-DI-HEA found in Venus Diamond (Kulzer), the dimer acid monomers used in N'Durance (Septodont) have been shown to have lower polymerization shrinkage than bis-GMA-based materials [28-30].

A new group of resin composites enable bulk placement came recently available. They can be divided into two groups with different mechanical properties, the low- and high-viscosity materials. As opposed to the high viscosity materials, those with low viscosity must be covered with an oclussal layer of a conventional hybrid resin composite. For the first marketed flowable bulk-fill composite resin SDR ('Smart Dentin Replacement', Dentsply DeTrey) polymerization stress was claimed to be reduced directly during curing. A polymerization modulator, a patented urethane di-methacrylate, is chemically embedded in the resin backbone, which results in a slower modulus development, allowing stress reduction without decreasing conversion rate [31].

In spite of the fact that the development of low shrinkage and low shringkage stress resin composites shows significant progress in last decade, it is still far away from non shrinking and non stress developing restorative materials.

1.6 Properties of Silorane-based composite

Expanding monomer systems of the double ring - opening polymerization of bicyclic monomers were first reported by Bailey in 1972 [32]. Since this time several kinds of ring-opening monomers were synthetized and tested for dental use or incorporated in formulations with conventional methacrylate monomers to decrease or eliminate polymerization shrinkage [33-35]. However, all these developments are experimental at the present moment.

Siloranes are the first commercially available resin composites based on ring opening monomer system. The term 'Silorane' derives from its chemical building blocks; siloxanes and oxiranes [36]. Cyclosiloxanes are responsible for the high hydrophobicity [37] of the material while the cycloaliphatic oxiranes guarantee reactivity. Silorane resin reveals lower polymerization shrinkage compared to the dimethacrylates. The polymerization shrinkage of 'Filtek Silorane' is claimed to be about 1 vol%. In contrast to methacrylates, which polymerize through radical addition reaction of their double bonds, siloranes polymerize through cationic ring opening reaction. The cationic cure starts with the initiation process of an acidic cation which opens the oxirane ring and generates a new acidic center, a carbo-cation. After the addition to an oxirane monomer, the epoxy ring is opened to form a chain, or in the case of two- or multifunctional monomers a network is formed [36,38]. Studies on biocompatibility and cytotoxycity of siloranes showed similar or slightly better results than methacrylates [39] and their mechanical properties seems to be equivalent to methacrylate based materials [40].

The new chemistry makes siloranes incompatible with methacrylate-based adhesive systems. Therefore, Filtek Silorane comes with a dedicated two-step self-etch adhesive, called 'Silorane System Adhesive' (3M-ESPE, USA). In contrast to most 2-step self-etching adhesive systems, in which only the bond is light cured (the primer is usually not), the SSA-Primer requires polymerization before the application of the bonding layer. Regarding to the fact that both layers i.e. SSA-Primer and SSA-Bond have to be polymerized separately, SSA-Primer can be categorized as a one-step self-etch adhesive system and SSA-Bond as a hydrophobic viscous coating resin establishing the compatibility between the hydrophilic SSA-Primer and the hydrophobic Silorane composite [36].

1.7 Marginal integrity testing

Marginal adaptation is one of the factors of the United States Public Health Service (USPHS) criteria together with retention, staining, marginal discoloration, surface roughness and sensitivity that is used in most clinical studies to judge on the restoration's clinical success [41]. *In vitro* evaluation of marginal adaptation is based on the fact that by identifying defects at the tooth-restoration interface, an early sign of adhesive failure is already affecting the restoration before catastrophic failures like restoration loss can occur.

While rather simple tests were initially applied to the resin composite restorations such as dye, isotope or bacteria infiltration tests, much more sophisticated ones including eventually thermal and mechanical loading are nowadays applied [42,43]. Scanning electron microscopy (SEM) and quantitative marginal assessment proved to be complementary evaluation methods. SEM analysis provides the microscopic details of the continuity of resin-enamel and resin-dentin interface and marginal analysis allows the quantification of the rate of continuous gap-free margins on both tooth interfaces. This technique is nondestructive as by analyzing gold-coated replicas, marginal qualities can be assessed both before and after loading [44].

Despite a recent review [45] questioning the relevance of marginal-integrity tests, it must be admitted that phenomena such as microleakage, pulpal complications, secondary caries and fractures, which are induced by interface breakdown represents the majority of all clinical failures observed in all types of direct restorations [46]. SEM analysis of the adhesive interface complemented by Optical Coherence Tomography confirmed that the presence of marginal gaps could be considered as an early sign of adhesive failure that on the long term, led to restoration loss if more than 50% of marginal openings were detected on enamel and dentin margins of Class V restorations [47]. In a recent study, a high correlation was observed between clinical and laboratory data of marginal adaptation provided that the same restorative material is considered in both *in vitro* and *in vivo* studies [48]. Therefore, the clinical behaviour of restoration margins can be predicted on the basis of *in vitro* tests on marginal integrity, as also shown by Frankenberger and coworkers [49]

1.8 Aim of the thesis

The general purpose of this thesis is to evaluate selective properties of a low shrinking silorane-based resin composite and to compare them with commonly used methacrylate-based materials. This thesis mainly focuses on the influence of the low polymerization shrinkage and shrinkage stress formation on the marginal adaptation. The other factors such as different adhesive application protocols, different adhesive system combinations and dentinal fluid simulation in relation to the marginal adaptation are studied as well. Furthermore, the staining susceptibility of a Silorane resin composite is evaluated.

The first specific aim of this thesis is to investigate the influence of volumetric shrinkage and C-factor on marginal adaptation of Class I composite restorations. The shrinkage kinetics will be evaluated using the linear displacement (LD) and shrinkage force (SF) measurements. Then Class I cavities of different the C-factor simulated by using total or selective bonding application will be restored with Silorane and methacrylate-based composites. This study is described in **Chapter 2**.

In **Chapter 3**, the influence of different bonding strategies on the marginal and internal adaptation of Class I Silorane restoration will be evaluated. In contrast to most 2-step self-etch adhesive systems Silorane System Adhesive (SSA) requires primer polymerization before application of the bonding layer. Additional enamel etching, selective bonding application and omitting of SSA-Primer polymerization will be compared to standard SSA application.

In **Chapter 4**, the effect of different one-sep self-etch adhesives on the marginal adaptation of Silorane and methacrylate-based composite in Class V cavities will be studied. As the SSA-Primer from Silorane System Adhesive has to be polymerized, it can be categorized as one-step self-etch system. This adhesive will be replaced by different one-step self-etch adhesives following application of SSA-Bond. The Class V cavities will be thereafter restored either with Silorane or methacrylate composite.

In **Chapter 5**, the the aim is to evaluate the effect of dentinal fluid simulation on the marginal adaptation of Silorane composite in Class I and Class V cavities. The dentinal fluid simulation will be performed during composite placement, polishing and thermo-mechanical loading. The results of marginal adaptation in Class I and Class V cavities done without dentinal simulation will be used as a control.

The in-vitro trials presented in Chapters 2-5 were standardized in an effort to simulate oral environment. Natural teeth, dentinal pulp simulation and moist environment were maintained. The teeth were loaded in computer-controlled chewing machine. Thermal and mechanical loading was applied simultaneously. Thermal cycling was performed in flushing water with temperatures changing 3.000x from 5ºC to 50ºC, and the mechanical loading was performed with $1.2x10^6$ load cycles transferred to the center of the occlusal surface at a frequency of 1.7Hz. A maximal load of 49N is applied by using a natural lingual cusp taken from extracted human molars.

The last specific aim of the thesis is to investigate long-term staining susceptibility of chemically different based composites using two different color-difference formulas. The Silorane together with ormocer, methacrylate and compomer composite will be immersed for 99 days in 5 staining solutions (red wine, juice, coke, tea and coffee). Spectrophotometric measurements will be done before and after staining and color changes determine according to ΔE and ΔE_{00} formula. This study is described in **Chapter 6**. Finally the thesis is summarized in English and Dutch

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Chapter 2

Shrinkage kinetics of a methacrylate and a silorane based resin composite: effect on marginal integrity

This chapter is published in Journal of Adhesive Dentistry 2013: Ladislav Gregor, Tissiana Bortolotto, Albert J. Feilzer, Ivo Krejci. Shrinkage kinetics of a methacrylate and a silorane based composite resin: effect on marginal integrity.

2.1 Abstract

Objectives: The aim of this study was to evaluate the relation between the linear displacement (LD), shrinkage force (SF) and marginal adaptation of a methacrylate- and a silorane-based composite.

Methods: LD and SF of 8 samples made out of Filtek Supreme XT (methacrylate-based composite) and Filtek Silorane (silorane-based composite) were measured for 180 s from the start of polymerization. Large bulk filled Class I cavities were restored with both resin composites, and two C-factors were simulated by applying the adhesive system in different ways: Silorane System Adhesive (SSA) was applied on enamel and dentin (C-factor 3.5) or only on enamel margins (C-factor of 0.4). Percentages of continuous margins (%CM) were quantitatively assessed on SEM before and after loading with 1.2 million mechanical occlusal cycles (49 N; 1.7 Hz) and simultaneous 3000 thermal cycles (5-50°C) under dentinal fluid simulation.

Results: Significantly lower scores of LD and SF were observed for Filtek Silorane (LD: 12.0 ± 1.3 µm, SF: 13.7 ± 1.0 N) than for Filtek Supreme XT (LD: 25.0 ± 0.6 µm, SF: 36.3 ± 2.9 N). Both variables, i.e. composite type and C-factor had a significant effect on marginal adaptation ($p < 0.05$). In the groups with high C-factor (SSA was applied on the entire cavity surface) %CM (mean ± SD) before / after loading, respectively, was 23 ± 4.9 % and 1.9 ± 0.7 % for Filtek Supreme XT, and 62.5 ± 8.9 % and 40.3 ± 7.1 % for Filtek Silorane. When adhesion was confined to enamel margins (lower C-factor), %CM before and after loading, respectively, increased to 76.1 ± 9.6 % and 64.2 ± 11.5 % for Filtek Supreme XT, and 96.6 ± 1.7 % and 94.2 ± 2.2 % for Filtek Silorane.

Conclusions: The silorane-based composite exhibited significantly lower shrinkage forces and better marginal adaptation than methacrylate-based one.

2.2 Introduction

Polymerization shrinkage is one of the major problems associated with direct resin bonded composite restorations (RBCs) [1]. The majority of commercially available dental resin composite materials are based on dimethacrylate monomers such as Bis-GMA or UDMA. During the free-radical polymerization the conversion of methacrylate monomer molecules into a polymer network results in a closer packing of the molecules leading to volumetric shrinkage [2]. This polymerization shrinkage creates contraction stresses that build up at the interface between the restorative material and cavity walls [3,4]. Polymerization shrinkage stress may cause deformation of the tooth [5], open pre-existing or create new enamel microcracks [6,7] and even initiate microcracking within the restorative material [8]. On the composite/tooth interface, polymerization shrinkage stress can lead to adhesive failures which may cause microleakage, marginal discoloration and post-operative sensitivity [9]. Various incremental layering techniques [10], application of low-modulus intermediate layers [11], use of different light curing protocols [12] or modification of the C-factor [13] have been described in the literature to reduce the effects of the inherent polymerization shrinkage stress. However, that strategies are often difficult to perform, time consuming and technique sensitive. This is why research focuses on the development of low or even zero shrinking materials, which would allow for simpler, faster and more reliable restorative techniques.

Expanding monomer system of the double ring-opening polymerization of bi-cyclic monomers was first reported by Bailey [14]. Since then, several kinds of ring opening monomers have been synthesized and tested for dental use separately or incorporated in formulations with methacrylate monomers to reduce or eliminate polymerization shrinkage. However, all these developments have been performed under experimental conditions. Siloranes are the first commercially available resin composites based on a ring opening monomer system. The term "silorane" derives from its chemical building blocks of siloxanes and oxiranes [15]. Cyclosiloxanes are responsible for the high hydrophobicity [16] of the material while the cycloaliphatic oxiranes guarantee reactivity [17]. In contrast to methacrylates which polymerize through radical addition reaction of their double bonds, siloranes polymerize through cationic ring opening reaction, thus reducing polymerization shrinkage [17]. Different silorane chemistry makes them not compatible with methacrylate adhesive systems [15,17].

Using Filtek Silorane as restorative material requires the application of a dedicated Silorane System Adhesive (SSA) consisting of the self-etch primer (SSA-Primer) and bond (SSA-bond). Duarte *et al.* [18] demonstrated acceptable bond strengths between hydrophobic SSA-bond resin and conventional total-etch methacrylate adhesive. Tzvergil-Mutulay *et al.* [19] also reported a good adhesion between SSA-bond and dimethacrylate based resin composite. However, in addition to the strength of the bond, shrinkage kinetics of the composite material during polymerization has also an influence on the quality of adhesion. Feilzer and others [13] showed that shrinkage stress is related to the configuration factor (C-factor), defined as the ratio of bonded to unbonded surfaces of the restoration. It is well known that in cavities with a low C-factor (less than 1), higher potential remains for plastic deformation and thus relaxation of the material, resulting in lower shrinkage stress [13]. Interestingly, the effect of a low-shrinking resin composite (eg, siloranes) vs. a methacrylate-based resin composite on marginal adaptation is poorly reported in the literature.

The purpose of this *in vitro* study was to evaluate the effect of polymerization shrinkage and thermomechanical loading on marginal adaptation of two different composites used to restore Class I cavities with a high or a low C-factor using the same adhesive system. Therefore, the linear polymerization displacement and polymerization shrinkage force of a Silorane- and a methacrylate-based resin composite was also determined.

The hypotheses tested were: first, that two composite materials with different chemical composition would behave distinctly in terms of polymerization shrinkage, second, that the composite with lower shrinkage properties would perform better in terms of marginal adaptation before and after thermo-mechanical loading, and third, that a low C-factor (which was simulated by avoiding adhesion to dentin) would positively influence the quality of marginal adaptation.

2.3 Materials and Methods

Polymerization shrinkage

Measurements of the linear displacement induced by polymerization shrinkage was performed with a custom made measuring device previously described in detail by Stavridakis *et al.* [20] In short, it consisted of a stable metal frame, upon which a thin aluminium platelet with a perpendicular diaphragm extended into a recess in the measuring sensor. The displacement

of the aluminium platelet caused by polymerization shrinkage of the test material was detected by a temperature compensated infrared sensor with an accuracy of 100 nm and a sampling frequency of 1 Hz. The data were recorded by means of an A/D converter using custom made software with a personal computer (Macintosh IIfx; Apple computer, Cupertino, CA, USA).
Filtek Supreme XT (LOT:7CF) and Filtek Silorane (LOT: 203905) were tested. Light polymerization was carried out for 60 s (L.E.Demetron II, Serial No: 792026758, Kerr, Orange, CA, USA) with a intensity of 800 mW/cm^2 (Curing Radiometer, Demetron Research, Danbury, CT, USA). Eight measurements were carried out on each tested material and their mean values at 180 s after the start of the polymerization were calculated.

Measurements of polymerization shrinkage force were performed with a custom made measuring device that was also previously described in detail by Stavridakis *et al.* [20]. Briefly, the upper part of the apparatus consisted of a semirigid load cell (PM 11-K; Mettler, Greifensee, Switzerland), to which a metal cylinder was screwed to mimic the natural deformation of cavity walls. The cylinder was coated with a standardized amount of composite which was compressed at a thickness of 1.5 mm onto a glass plate attached to the base of the device. The surfaces of the metal cylinder and of the glass plate were sandblasted with 50 μm Al_2O_3 (Microetcher; Danville Engineering, Danville, CA, USA) and silanized (Monobond S; Ivoclar Vivadent). The force that built up during polymerization shrinkage was detected by means of load cell at a sampling frequency of 1 Hz. The data were transferred real time into attached computer (Macintosh IIfx; Apple computer, Cupertino, CA, USA) via an A/D converter using custom made software. The same materials, the same light curing unit and the same number of samples per group were used as for the linear displacement. The use of 180 s as a standard analysis time was chosen according to the results of Stavridakis *et al.* [20], showing that for both linear displacement and shrinkage force measurements, the results after 180 s attain over 90% of the maximum values after 20 min.

Marginal adaptation

Forty caries free human lower third molars with completed root formation were stored in 0.1% thymol solution until their use for the experiment. After cleaning, the apices were sealed using an adhesive system (Optibond FL, Kerr, Orange, CA, USA) and the teeth were mounted in the centre of custom made specimen holders using a cold-polymerizing resin (Technovit 4071, Heraeus Kulzer GmbH, Wehrheim, Germany). All teeth were prepared for the simulation of dentinal fluid using horse serum diluted to 1:3 ratio with 0.9% NaCl under hydrostatic pressure of about 25 mm Hg [21]. The intrapulpal pressure was maintained during cavity preparation, restoration placement and thermo-mechanical loading.

Standardized, beveled, model Class I cavities were prepared under copious water spray cooling by using 80 μm diamond burs and 40 μm finishing diamond burs. The dimensions of each cavity was 5.0 ± 0.5 mm vestibulo-lingually, 7.0 ± 0.5 mm, mesio-distally, and 2.5 ± 0.5 mm in depth. Each bur was replaced with a new one after four cavity preparations. All the teeth were randomly divided into 4 groups (A to D) (n=10). Silorane System Adhesive (SSA, LOT: 7AA, 3M-ESPE AG, Seefeld, Germany) was applied according to manufacturer's instructions in groups A and B. Both SSA-primer and SSA-bond were polymerized for 10 s (L.E.Demetron II, Kerr, Orange, CA, USA). Then the teeth were restored with a methacrylate-based resin composite (Filtek Supreme XT, LOT: 7CF, 3M-ESPE, St. Paul, USA) or with a silorane composite (Filtek Silorane, LOT: 7AJ, 3M-ESPE, St. Paul, USA) using a simple bulk technique, then cured from the occlusal for 60 s (L.E.Demetron II). To simulate a Class I cavity with a low C-factor, SSA-bond was applied only on etched enamel (37% H_3PO_4) for 30 s and polymerized for 10 s (L.E. Demetron II) in groups C and D, while dentin adhesion was intentionally omitted, ie, adhesion was confined to enamel margins. Then the teeth were restored using the same composite materials, placement technique and light curing protocol as in groups A and B (total bonding concept). Immediately after polymerization, the restorations were finished with fine diamond burs (Intensiv SA, Grancia, Switzerland) and polished with flexible aluminum oxide discs (Sof-Lex Pop-On, 3M-ESPE, St. Paul, MN, USA).

After storage for one week in water at 37°C in the dark, the restored teeth were loaded in a computer-controlled chewing machine. Thermal and mechanical loads were applied simultaneously [22,23]. Thermal cycling was performed in flushing water with temperatures changing 3,000x from 5°C to 50°C, the mechanical loading performed with 1.2 million load cycles transferred to the center of the occlusal surface at a frequency of 1.7 Hz. A maximal load of 49 N was applied by using a natural lingual cusp taken from extracted human molars. Replicas of each restoration before and after loading were readied by using a polyvinylsiloxane material (President light body, Coltène-Whaledent AG, Altstätten, Switzerland). Gold-coated epoxy replicas were prepared for the computer assisted quantitative margin analysis in a scanning electron microscope (SEM; XL20, Philips, Eindhoven, the

Netherlands) and evaluated at 200x magnification. The marginal quality was expressed as percentages of "continuous margins" present.

Statistical analysis

Statistical analysis was performed with SPSS 16.0 for Windows. Normal distribution of marginal adaptation data (Kolmogornov-Smirnov) allowed the use of a one-way ANOVA. The post hoc Duncan test helped to identify differences between groups. For each group, a paired t-test was used to detect whether the differences between and after loading were significant or not. Results of linear displacement and polymerization shrinkage force were analyzed with an unpaired simple t-test. The confidence level was set to 95%.

2.4 Results

Figures 2.1 and 2.2 illustrate the shrinkage development over time for both composite resins. Means ± SD of linear displacement after 180 s were 25.0 ± 0.6 µm for Filtek Supreme XT and 12.0 ± 1.3 µm for Filtek Silorane. Polymerization shrinkage forces after 180 s amounted to 36.3 ± 2.9 N for Filtek Supreme XT and 13.7 ± 1.0 N for Filtek Silorane. The differences between materials were significant, both for linear displacement and for polymerization shrinkage force (unpaired t-test, $p<0.05$).

Means ± SD of percentages "continuous margins" (%CM) before/after loading were of 24.4 ± 16.6 % / 2.1 ± 2.4 % for Filtek Supreme XT and 58.8 ± 9.9 % / 35.4 ± 4.1 % for Filtek Silorane in cavities with high C-factor (Fig 2.3) and 76.1 ± 9.6 % / 64.2 ± 11.5 % for Filtek Supreme XT and 96.6 ± 1.7 % / 94.2 ± 2.1 % for Filtek Silorane in cavities with low C-factor (Fig. 2.4). The differences between materials were significant both before and after loading ($p<0.05$). A significantly higher %CM was observed in both groups when the low C-factor was simulated ($p<0.05$), indicating that when adhesion was established on enamel only, the performance of both restorative materials was significantly higher. A significant marginal degradation from before to after loading was observed in all groups, with the exception of the Silorane group with low C-factor (96.6 ± 1.7 %) before loading, (94.2 ± 2.1 %) after loading.

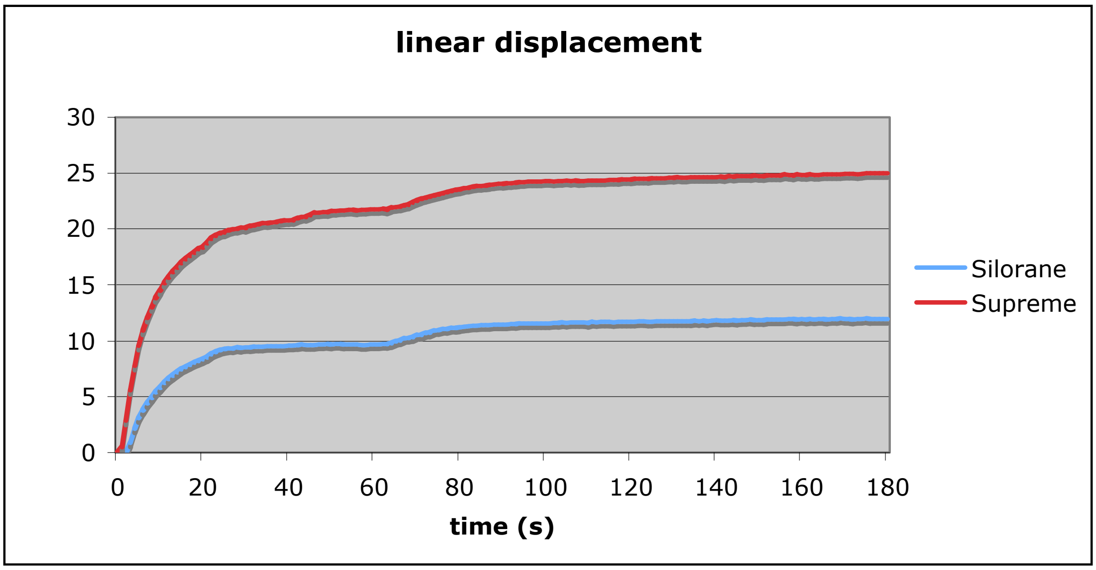

Fig. 2.1: Mean linear displacement (µm) curves of the two composite materials over a period of 180 s. The slight increase in shrinkage at 60 s is due to the thermal effect of the light-curing unit (2 mm thick resin composite sample polymerized with L.E. Demetron II).

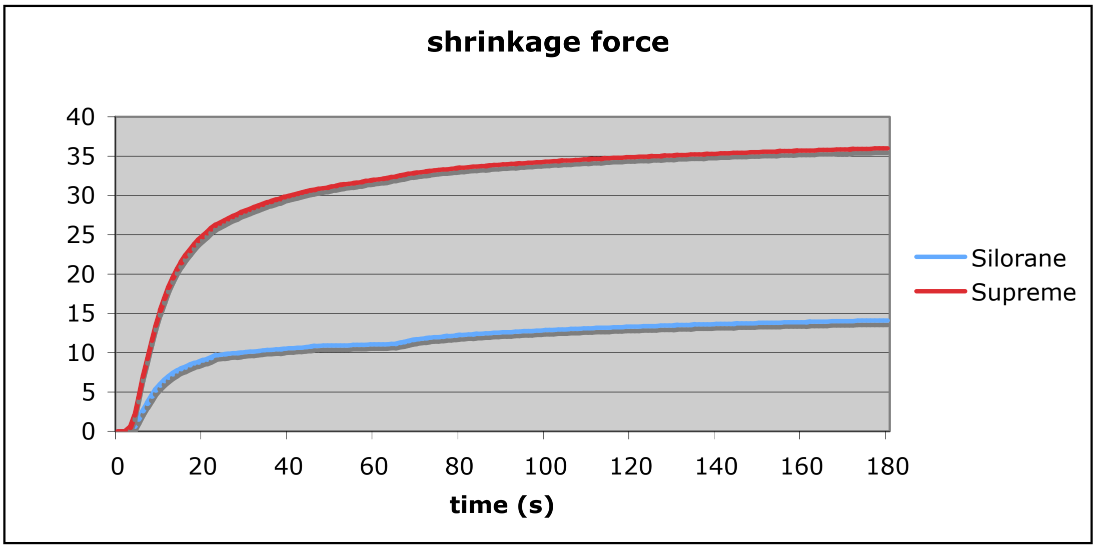

Fig. 2.2: Mean polymerization shrinkage force (N) curves of the two resin composite materials over a period of 180s. The slight increase in shrinkage at 60 s is due to the thermal effect of the light-curing unit (L.E. Demetron II).

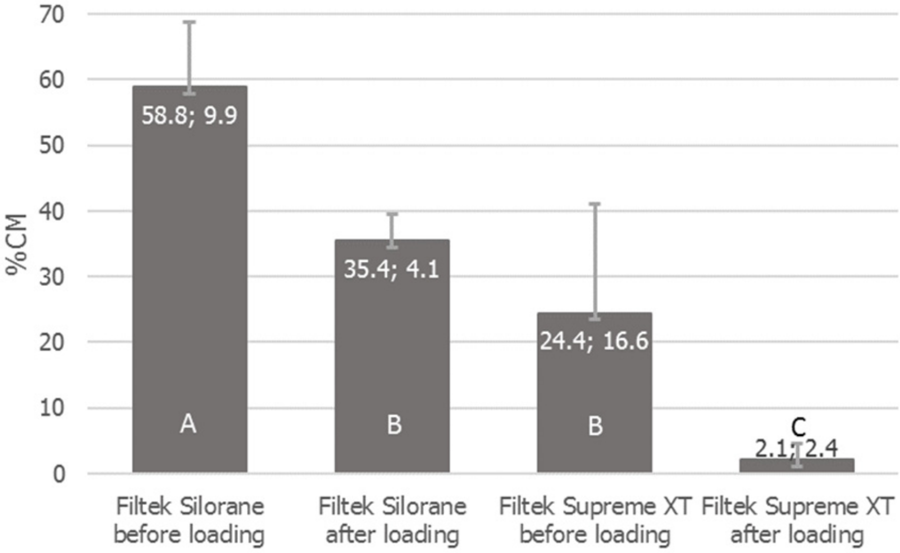

Fig. 2.3: Results of marginal adaptation in cavities with high C-Factor. Different letters indicate significant differences between groups ($p<0.05$).

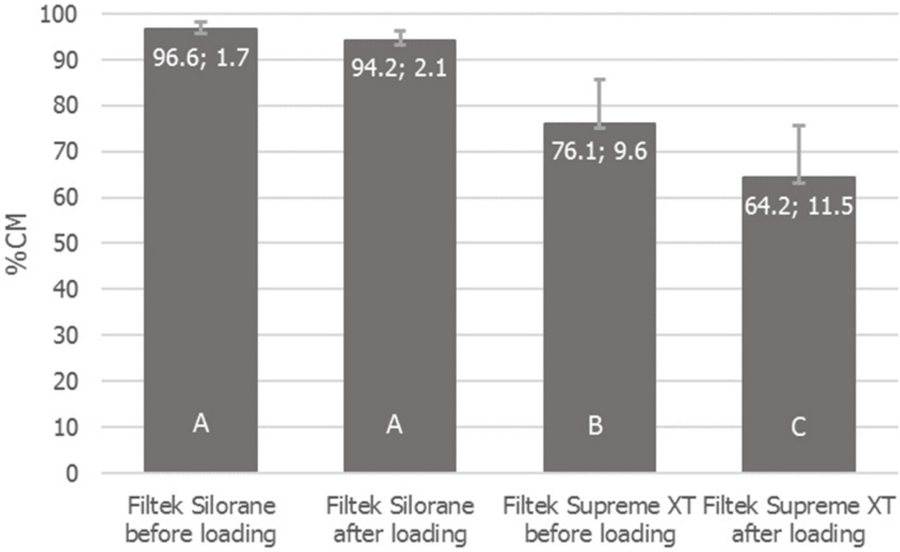

Fig. 2.4: Results of marginal adaptation in cavities with low C-Factor. Different letters indicate significant differences between groups ($p<0.05$).

2.5 Discussion

Since marginal adaptation has been described as one of the most important factors that influence the clinical outcome of an adhesive restoration [24], this study evaluated the influence of polymerization shrinkage and polymerization shrinkage force of a silorane-based composite in comparison to a methacrylate-based composite on marginal adaptation and fatigue resistance in a Class I cavity, where the same adhesive system was applied in cavities with two different C-factors (high and low). According to the "total-bonding" concept, an adhesive system composed of a primer and a bond must be applied on both enamel and dentin substrate. This procedure was followed in our protocol (groups A and B), meaning that the adhesive system consisting of SSA-Primer and SSA-Bond were applied on both enamel and dentin before the insertion of the resin composite. In order to observe the behavior of both composites in the cavities with a low C-factor, an additional evaluation (groups C and D) was performed where the adhesion was confined to enamel margins. In this way, the C-factor was modified while keeping constant the type of cavity (Class I). In fact, SSA-Primer was omitted and SSA-Bond was applied only on etched enamel; the C-factor for this type of cavity was 0.4 *vs*. 3.5 [13] when the total-bonding concept was used. A chewing machine comprising thermocycling and cyclic occlusal loading together with the simulation of dentinal fluid was used to fatigue the specimens [21,25].

Polymerization stress development is a complex process involving several factors, such as volumetric shrinkage, curing rate, viscoelastic behavior of the composite, bonding capacity of the adhesive system, and C-factor of the cavity [4,26-28]. To assess the effect of polymerization shrinkage on marginal adaptation, the same adhesive system, curing protocol, and cavity type were used in all groups. The Class I cavities were filled in bulk to increase contraction stresses generated during light polymerization. By this, the most clinically unfavorable situation in terms of shrinkage stress development was simulated. A maximum cure rate was achieved by light curing with an energy density of 48 J/cm^2 (60 s x 800mW/cm^2). Earlier studies showed that optimal polymerization occurred already at energy densities of 16 J/cm^2 [29-33]. The methodology for the linear displacement and polymerization shrinkage force measurement used in this experiment was based on the developments of De Gee *et al.* [34]. As proposed by Stavridakis *et al.* [34], the linear displacement measured in this study was reported in µm, as this was the recording unit of the infrared sensor that was used for measuring the vertical displacement of the diaphragm caused by polymerization shrinkage. Linear polymerization shrinkage may be calculated using the following equation according to De Gee *et al.* [34]:

$$lin\,\% = \frac{\Delta L}{L + \Delta L} \times 100$$

where ΔL is the recorded displacement and L the thickness of the sample after polymerization. Using this formula, data recorded under the conditions of this study correspond to the following values: 0.80% for Filtek Silorane and 1.65% for Filtek Supreme XT for linear polymerization shrinkage, which is within the range of values reported by other researchers [15,20]. The results of polymerization shrinkage force are reported in Newtons. Taking into consideration the C-factor of 2.67 of specimens used in this experimental semi-rigid set-up, the measured forces are lower in comparison to the results reported by other researchers who used rigid experimental set-ups [20].

In respect to linear shrinkage and shrinkage force, Filtek Silorane performed significantly better in comparison to Filtek Supreme XT, confirming the reduced shrinkage of the silorane compound. Thus, the first hypothesis was accepted. In terms of marginal adaptation, Filtek Silorane performed significantly better than Filtek Supreme XT both before and after loading. Therefore, the second hypothesis was also accepted. Very low percentages of marginal adaptation before / after loading (23 ± 4.9 % / 1.9 ± 0.7 %) were attained by the methacrylate-based resin composite (Filtek Supreme XT). This could be partly explained by a higher polymerization shrinkage and shrinkage force in respect to the silorane-based material (Fig. 2.1, Fig. 2.2). Differences in E - moduli between Filtek Supreme XT and Filtek Silorane [35-38] may have also accounted for the results, as the extent of shrinkage stress is dependent on the viscoelastic properties of the resin composite. At a given polymerization shrinkage, a more rigid resin composite may be subject to higher shrinkage stress and, consequently, increase gap formation at the tooth-resin composite interface [26,39].

In the cavities with simulated low C-factor, the results of marginal adaptation on enamel were for both methacrylate-based and silorane-based composites significantly better (Fig. 2.4). Moreover, in the case of Silorane, loading did not significantly influence the quality of marginal adaptation. The third hypothesis was therefore accepted.

As opposed to most 2-step self-etching adhesive systems, in which only the bond is light cured (the primer is usually not), the SSA-Primer requires polymerization before the application of the bonding layer. SSA-Bond acts as a hydrophobic viscous coating resin and its main purpose is to establish the compatibility between the hydrophilic SSA-Primer and the hydrophobic silorane composite [15]. In the present study, the marginal imperfections observed on both methacrylate and silorane groups were located at the enamel/bond interface. No adhesive failures at the bond/composite interface were detected, proving that SSA-Bond is compatible with both methacrylate and silorane-based composite resins. Our observations support those of recent studies [18,19,40] in the sense, that SSA-Bond is, in fact, compatible with methacrylates.

2.6 Conclusions

When comparing a methacrylate and silorane-based composite resin in terms of marginal adaptation and shrinkage development, the superior results observed with the silorane material could be attributed to its lower polymerization shrinkage stress development. From a clinical standpoint, our results show that methacrylate-based composites can be safely combined with the Silorane System.

2.7 References

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Chapter 3

Effect of different bonding strategies on the marginal adaptation of Class I silorane restoration

This chapter is published in American Journal of Dentistry 2013: Ladislav Gregor, Tissiana Bortolotto, Albert J. Feilzer, Ivo Krejci. Effect of different bonding strategies on the marginal adaptation of Class I silorane restoration.

3.1 Abstract

Objectives: To evaluate the quality of marginal and internal adaptation of Filtek Silorane composite in standardized Class I cavities before and after thermo-mechanical loading using different application protocols of the Silorane System Adhesive (SSA).

Methods: Five groups (n=10) of Class I cavities were restored with Filtek Silorane using different SSA applications. Total bonding (TB): Group A (SSA), Group B (SSA without primer polymerization), Group C (enamel etching + SSA), Group D (enamel etching + SSA without primer polymerization) and Selective bonding (SB): Group E. Marginal adaptation was assessed on replicas in the SEM at 200x magnification before and after thermo-mechanical loading (3,000 × 5-55°C, $1.2 \cdot 10^6 \times 49$ N; 1.7 Hz) under simulated dentinal fluid. After loading the samples were sectioned and the internal adaptation was evaluated as well.

Results: The lowest scores of %CM (Continuous Margin) before/after thermo-mechanical loading being 80.8 ± 8.2 % / 32.1 ± 8.3 % were observed in the control group A. Enamel phosphoric acid etching prior to the application of the SSA resulted in significantly higher %CM before and after loading in comparison with the "non-etched" groups ($p > 0.05$). When enamel etching was performed before the application of the adhesive system no statistically significant differences ($p > 0.05$) were observed regardless of how the SSA was applied (total vs. selective bonding). Internal adaptation was negatively influenced by omitting the SSA-Primer polymerization ($p > 0.05$).

Conclusions: Etching enamel with H_3PO_4 prior to SSA application significantly improve the marginal adaptation of Silorane composite. As the non-polymerization of SSA-Primer polymerization negatively influences dentinal adhesion it is mandatory to polymerize the SSA-Primer.

3.2 Introduction

Polymerization shrinkage still remains a major drawback of dimethacrylate resin composite materials [1]. Volumetric shrinkage is clinically undesirable because it may stress the adhesive interface and cause tooth deformation [2], microcracking within the bulk of the composite [3,4], enamel microcracks [5] or microleakage [6].

Recently, a new class of low-shrinking composites based on silorane technology (Filtek Silorane, 3M ESPE, Seefeld, Germany) with the volumetric shrinkage of about 1% [7] was introduced to the dental profession. The low shrinkage is due to ring opening polymerization of the silorane molecules instead of free radical polymerization of methacrylate monomers. Studies on cytotoxicity [8] and mutagenity [9] of siloranes showed similar or slightly better results than methacrylates and their mechanical properties seem to be equivalent to methacrylate based materials [10-13].

The new chemistry makes siloranes incompatible with methacrylate-based adhesive systems [7,14]. Therefore Filtek Silorane comes with a dedicated two-step self-etch adhesive, called Silorane System Adhesive (3M ESPE, USA). In contrast to most 2-step self-etching adhesive systems, in which only the bond is light cured (the primer is usually not), the SSA-Primer requires polymerization before the application of the bonding layer [7]. Regarding to the fact that both layers i.e. SSA-Primer and SSA-Bond have to be polymerized separately, SSA-Primer can be categorized as a one-step self etch adhesive system and SSA-Bond as a hydrophobic viscous coating resin establishing the compatibility between the hydrophilic SSA-Primer and the hydrophobic silorane composite.

Previous laboratory studies proved the benefit of Filtek Silorane low polymerization shrinkage on marginal adaptation and microleakage formation in comparison to methacrylate based [15-17] and to ormocer based [17-18] composites. However in prospective randomized clinical trial Schmidt *et al.* [19] observed slightly inferior results of marginal adaptation of Filtek Silorane in comparison with CeramX after 1 year of clinical service.

Several strategies of methacrylate based self-etch adhesive systems' application have been described in the literature to enhance bond strength between methacrylate-based composites and dental tissues. Additional enamel etching [20,21] and special application techniques like selective bonding [22] multiple adhesive application or extra application of hydrophobic layer [23,24] have been described and evaluated in an effort to improve the seal of adhesive restorations.

It can be speculated, that similar to the results of the above mentioned studies, different strategies of SSA application may have a positive effect on marginal adaptation of a silorane-based resin composite. Therefore, the purpose of this laboratory study was to evaluate the effect of different application techniques of the Silorane System Adhesive on marginal and internal adaptation of Filtek Silorane composite class I cavities with a high C-factor before and after thermo-mechanical loading under the simulation of dentinal fluid. The null hypothesis tested was that there was no dofference in terms of marginal and internal adaptation when comparing different SSA application protocols in combination with low shrinkage silorane composite.

3.3 Materials and methods

Fifty caries-free human lower third molars with completed root formation were stored in distilled water until their use for the experiment. After cleaning, the apices were sealed using an adhesive system (Optibond FL, Kerr, Orange, CA, USA) and the teeth were mounted in the centre of custom made specimen holders using a cold-polymerizing resin (Technovit 4071, Heraeus Kulzer GmbH, Wehrheim, Germany). All teeth were prepared for the simulation of dentinal fluid using horse serum diluted to 1:3 ratio with 0.9% NaCl under hydrostatic pressure of about 25mmHg. The intrapulpal pressure was maintained during cavity preparation, restoration placement and thermo-mechanical loading [25]. The teeth were randomly distributed into five equal groups (n=10). Standardized Class I cavities with beveled margins (45° - 0.5 mm) were prepared in Groups A to D (total bonding) under copious water spray cooling by using 80 μm diamond burs and 40 μm finishing diamond burs. Enamel beveling was incorporated to the adhesive system application protocol in Group E (selective bonding) so the initial cavity preparation was done without email beveling. The dimensions of each cavity were 5.0 ± 0.5 mm vestibulo-lingual, 7.0 ± 0.5 mm mesio-distal and 2.5 ± 0.5 mm in depth. Each bur was replaced with a new one after four cavity preparations. The Silorane System Adhesive (Silorane System Adhesive, LOT: 7AA, 3M-ESPE AG, Seefeld, Germany) was applied to the cavities according to the adhesive protocol described in Table 3.1. The cavities were restored with Filtek Silorane composite (Filtek Silorane, LOT: 7AJ, 3M-ESPE, St. Paul, USA) (L.E.Demetron II, Serial No: 792026758, Kerr, Orange, CA, USA) using a two horizontal layer technique with a thickness of 1.5 mm each and polymerized for 20 s per layer (L.E.Demetron II, Serial No: 792026758, Kerr, Orange, CA, USA) with a power density of 1200 mW/cm^2. Immediately after polymerization, the restorations were finished by the use of fine diamond burs and polished by using flexible aluminum oxide discs with decreasing granulometry (Sof-Lex Pop-On, 3M-ESPE, St. Paul, MN, USA).

After storage for one week in water at 37°C in the dark, the restored teeth were loaded in a computer-controlled chewing machine. Thermal and mechanical loading was applied simultaneously. Thermal cycling was performed in flushing water with temperatures changing 3.000x from 5ºC to 50ºC, the mechanical loading performed with $1.2 \cdot 10^6$ load cycles transferred to the center of the occlusal surface at a frequency of 1.7 Hz. A maximal load of 49 N was applied by using a natural lingual cusp taken from extracted human molars [25,26].

Replicas of each restoration before and after loading were readied by using a polyvinylsiloxane material (President light body, Coltène-Whaledent AG, Altstätten, Switzerland). Gold-coated epoxy replicas were prepared for the computer assisted quantitative margin analysis in a scanning electron microscope (SEM; XL20, Philips, Eindhoven, the Netherlands) and evaluated at 200x magnification. The marginal quality was expressed as percentages of "continuous margins" before and after loading [25]. After the evaluation of marginal adaptation the samples were sectioned mesio-distally into two halves (buccal and lingual) using a slow rotating saw (Isomet,Buehlers) polished with flexible aluminium oxide discs (Sof-Lex Pop-On, 3M-ESPE, St. Paul, MN, USA) and the internal dentinal adaption was evaluated as well (Fig's 3.1-3.3) [25]. In an attempt to find out in which interface failures occurred, percentage of non-continuous margins were judged as either negative or positive interface failures. It was considered negative failure if detachment occurred between dentin (Fig. 3.2) and SSA and positive failure if the detachment occurred between SSA and silorane composite remaining the dentin unexposed (Fig. 3.3).

Table 3.1: Description of the application Silorane System adhesive tested

Group A	***total bonding*** Silorane System Adhesive: control group • *Silorane primer* application – scrubbing of the primer over the entire cavity for 15s followed by 10s polymerization • *Silorane bond* application and polymerization 10s
Group B	***total bonding*** Silorane System Adhesive: • *Silorane primer* application – scrubbing of the primer over the entire cavity for 15s ***primer polymerization omited*** • *Silorane bond* application and polymerization 10s
Group C	***total bonding*** 1) 30s selective enamel etching 36% H_3PO_4 2) Silorane System Adhesive: • *Silorane primer* application – scrubbing of the primer over the entire cavity for 15s followed by 10s polymerization • *Silorane bond* application and polymerization 10s
Group D	***total bonding*** 1) 30s selective enamel etching 36% H_3PO_4 2) Silorane System Adhesive: • *Silorane primer* application – scrubbing of the primer over the entire cavity for 15s ***primer polymerization omited*** • *Silorane bond* application and polymerization 10s
Group E	***selective bonding*** 1) Silorane System Adhesive: • *Silorane primer* application – scrubbing of the primer over the entire cavity for 15s followed by 10s polymerization • *Silorane bond* application and polymerization 10s 2) enamel preparation – enamel bewelling 3) 30s selective enamel etching 36% H_3PO_4 4) *Silorane bond* application and polymerization 10s

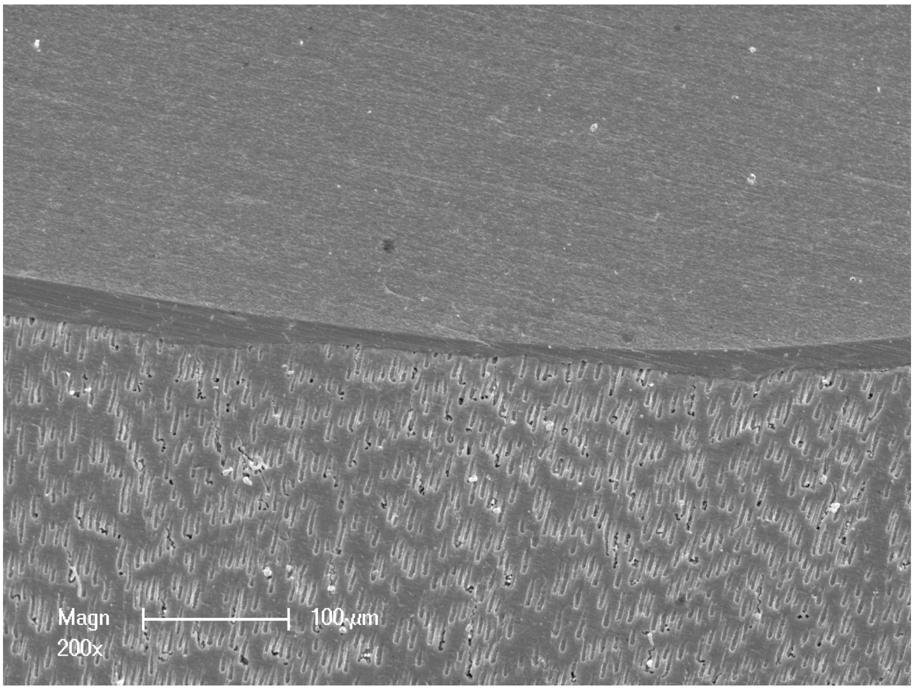

Fig. 3.1: Representative SEM micrograph of continuous margin

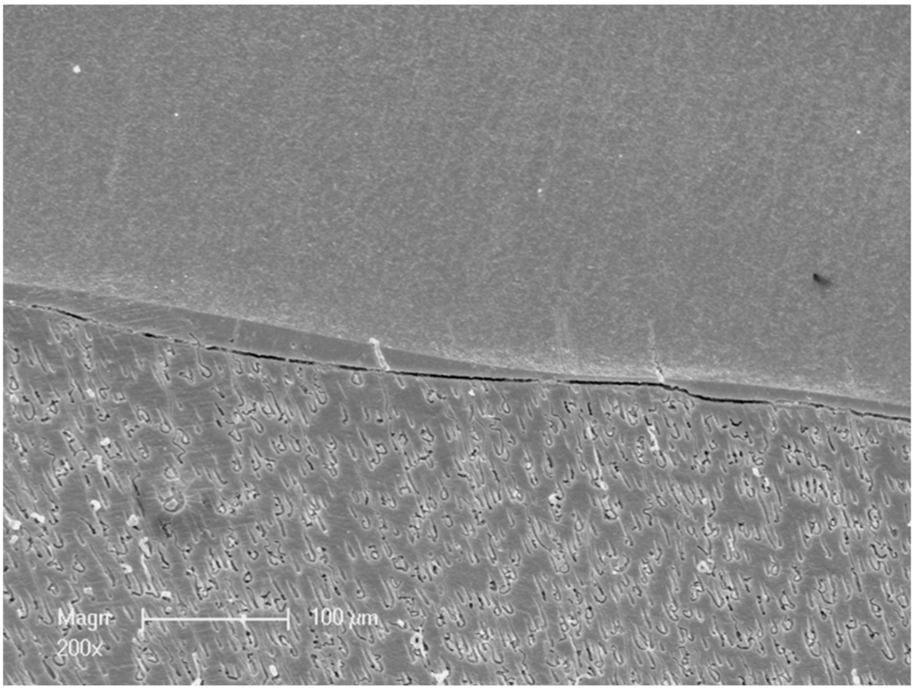

Fig. 3.2: Representative SEM micrograph of a "negative" failure between SSA and dentin

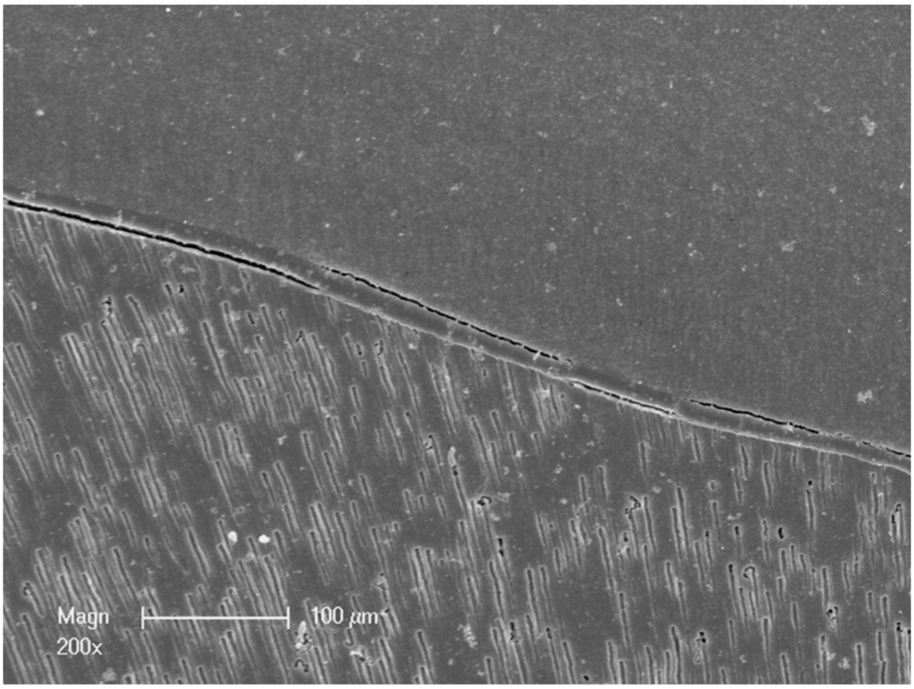

Fig. 3.3: Representative SEM micrograph of a different failures: "negative" between SSA and dentin and "positive" between SSA and silorane composite leaving the dentin sealed.

Statististics

Statistical analysis was performed with SPSS 16.0 for Windows. Normal distribution of the data both for marginal and internal adaptation (Kolmogornov-Smirnov) enabled the use of a one-way ANOVA. A post hoc Duncan test helped to identify differences between groups. The confidence level was set to 95%.

3.4 Results

The percentages of "continuous margins" (%CM) as well as the results of internal adaptation are presented in Table 3.2.

Marginal adaptation

Loading significantly influenced %CM in all groups ($p<0.05$). The lowest scores of %CM before/after thermo-mechanical loading (80.8 ± 8.2 % / 32.1 ± 8.3 %) were observed in the control Group A ("non-etched" group). Selective enamel phosphoric acid etching prior the application of the Silorane System Adhesive (Groups C, D) resulted in significantly higher %CM before and after loading in comparison with the "non-etched" groups ($p <0.05$). Omitting the

SSA-Primer polymerization did not result in decreased quality of marginal adaptation (p >0.05). When enamel etching was performed before the application of the adhesive system no statistically significant differences (p >0.05) were observed both before and after loading between the groups, regardless of how the Silorane System Adhesive was applied (total vs. selective bonding).

Table 3.2: Mean percentages (SD) of "continuous margin"

	Marginal adaptation			**Internal Adaptation**	
	Before loading	**After loading**		SSA-Silorane positive failure	Dentin-SSA negative failure
Group A	80.8 (8.2) **C**	32.1 (8.3) * **B**	90.8 (5.3) **A**	45%	55%
Group B	85.6 (6.1) **B**	38.3 (12.8) *__B__	67.8 (22.1) **B**	55%	45%
Group C	89.1 (6.3) **A**	68.5 (10.3) *__A__	88.8 (6.4) **A**	70%	30%
Group D	93.8 (4.5) **A**	70.8 (11.5) *__A__	71.3 (13.8) **B**	70%	30%
Group E	89.6 (10.1) **A**	74.1 (10.1) *__A__	85.3 (6.1) **A**	75%	25%

Group A – total bonding: SSA
Group B – total bonding: SSA without polymerization of SSA-Primer
Group C – total bonding: enamel etching + SSA
Group D – total bonding: enamel etching + SSA without polymerization of SSA-Primer
Group E – selective bonding: SSA, enamel beveling, enamel etching, SSA – Bond

(*) differences in mean values of marginal adaptation before and after loading in each group; applies to each line.
In capital letters (**A,B**): differences in mean values of marginal and internal adaptation between groups; applies to each column.

Internal adaptation

Internal adaptation was assessed after middle cutting the loaded specimens; therefore, results before loading are not presented. There were significantly better results ($p < 0.05$) observed in terms of internal adaptation in the groups where SSA-Bond and SSA-Primer were polymerized separately (Groups A, C and E). It was interesting to see that in groups where enamel was etched with H_3PO_4 (C, D and E), the failures predominantly occurred at the interface composite-adhesive system (positive failures). When enamel was conditioned only with the self-etching primer (Gr. A and B), a lower number of adhesive failures at the interface composite-adhesive system could be observed.

3.5 Discussion

This study evaluated the effect of different application techniques of Silorane System Adhesive on the marginal and internal adaptation. The chewing machine comprising thermal cycling and cyclic occlusal loading together with the simulation of the dentinal fluid was used for this purpose [25,27].

Bonding to phosphoric acid etched enamel is accepted as clinically strong and durable. The better results obtained with composites on enamel margins may be attributed to enamel composition and well-known bond stability of the adhesive systems to acid etched enamel [28-30]. SSA can be categorized as a two-step self-etch adhesive that bonds to enamel and dentin using a form of "nano-interaction", typical for 'ultra-mild' self-etch adhesives, and is related to the relatively high pH (2.7) of the SSA-Primer [31]. Even if self-etch adhesive systems are increasing in popularity because of user friendliness they might produce less durable bonds on enamel in comparison to etch and rinse adhesive systems [28-30].

The effect of enamel etching prior to the application of one-step self-etch adhesives was previously discussed in the literature. Higher μTSB [20,21] and better marginal adaptation [32] were demonstrated *in vitro* when one-step self-etch adhesives were used in combination with H_3PO_4 etching. Some studies showed that positive effect of H_3PO_4 etching might be product dependent [33,34]. More minor defects and restoration staining at the enamel margin were noticed *in vivo* when enamel was not selectively acid-etched [35].

The results of our study indicate that marginal adaptation of silorane composite is significantly influenced by enamel 37% H_3PO_4 etching. Therefore, the null hypothesis had to be rejected for the latter groups. Better results both before and after loading were observed if enamel was etched prior the SSA application. Taking into consideration the principle of marginal evaluation, i.e. marginal adaptation is expressed by the percentage of continuous margin (%CM), the quality of marginal seal dramatically decreased after loading in non-etched groups (A and B). This finding might be attributed to the assumption that SSA with relatively high pH (2.7) can provide less potential for micro-mechanical interlocking [30] which might result in lower bond durability [36]. Marginal adaptation was not significantly influenced by non-polymerization of the SSA-Primer. Compared to dentin, enamel contains less water and is probably more compatible with SSA bond.

Selective bonding technique as described by Krejci *et al.* [22] is a bonding technique where adhesion between the adhesive system and resin composite is not completely eliminated, but is considered weaker than the adhesion between the adhesive system and the tooth. The bonding potential of the adhesive is weakened by water contamination during the beveling of enamel margins. SSA selective bonding application resulted in better marginal adaptation compared to SSA total bonding approach. However, no differences in marginal adaptation were observed between SSA selective bonding (Group E) and the SSA total bonding using H_3PO_4 etching prior to the application of the Silorane System Adhesive (Group C). Our results support those of Schmidlin *et al.* [37] in high C-factor Class I cavities using a self-etch adhesive system showing no benefit of selective bonding on marginal adaptation compared to total bonding with enamel pre-etched with phosphoric acid.

In contras to marginal adaptation, internal (dentin) adaptation was affected when omitting SSA-Primer polymerization. Relatively high standard deviations and significantly lower scores of %CM were observed in the groups where SSA-Primer was not polymerized (B and D) confirming the need of SSA-Primer polymerization when bonding to dentin. The evaluation of internal adaptation showed high occurrence of failures close to the interface between the SSA and Silorane composite in all groups. The interface failures were judged as positive or negative as to whether dentin remained sealed or not. This evaluation followed the clinical point of view considering the possible presence of post-operative sensitivity. In groups where enamel was etched (C, D and E), the occurrence of positive failures mounted to 70%. This fact might be explained by the lower resistance of SSA/Silorane composite interface to increased internal stresses, when better marginal (enamel) adhesion is established [38]. Our findings are in agreement with the studies of Van Ende *et al.* [39] showing by μTBS-testing that on dentin, failures mostly occurred cohesively in the SSA layer or adhesively between the SSA and the Silorane composite suggesting that this interface is the weakest link.

In view of the fact that internal (dentin) adaptation showed high incidence of failures which occurred either cohesively in the adhesive layer or adhesively between the SSA and silorane composite leaving the dentin sealed, the present study concluded that the SSA total bonding application would behave similarly to a selective bonding technique at least from the micromorphological point of view. Additional clinical studies are necessary to validate this concept.

3.6 References

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Chapter 4

Marginal integrity of low shrinking versus methacrylate-based composite: effect of different one-step self-etch adhesives

This chapter published in Odontology 2016: Ladislav Gregor, Dorien Lefever, Tissiana Bortolotto, Albert J. Feilzer, Ivo Krejci. Marginal integrity of low shrinking versus methacrylate - based composite: effect of different one - step self - etch adhesives.

4.1 Abstract

Objective: The aim of the study was to evaluate the influence of composite type and adhesive system on the quality of marginal adaptation in standardized Class V cavities before and after thermo-mechanical loading (TML).

Methods: The cavities were restored using different combinations of three adhesive systems ((Silorane System Adhesive (SSA), Clearfil S^3 Bond (S3), G-Bond (G-B)) and two resin composite materials (Filtek Silorane, Clearfil AP-X). Six groups (n=10): Group A (SSA-Primer + SSA-Bond, Filtek Silorane) Group B (SSA-Primer + SSA-Bond, Clearfil AP-X) Group C (S3 + SSA-Bond, Filtek Silorane) Group D (S3 + SSA-Bond, Clearfil AP-X) Group E (G-B + SSA-Bond, Filtek Silorane) and Group F (G-B + SSA-Bond, Clearfil AP-X) were defined. Marginal adaptation was assessed on replicas in the SEM at 200x magnification before and after TML ($3{,}000 \times 5\text{-}55°C$, $1.2 \cdot 10^6 \times 49$ N; 1.7 Hz) under simulated dentinal fluid.

***Results*:** The highest scores of continuous margins (%CM) were observed in the group F (G-B + SSA-Bond, Clearfil AP-X: before loading 96.4 ± 3.2 % / after loading 90.8 ±7.0 %. A significant effect of adhesive system, composite type and loading interval was observed on the results ($p<0.05$). Significantly lower scores of %CM were observed for silorane-based composite (Filtek Silorane) after TML in comparison with methacrylate-based composite (Clearfil AP-X) considering total marginal length ($p<0.05$). For both Filtek Silorane and Clearfil AP-X, G-Bond performed significantly better than SSA-Primer and Clearfil S^3 Bond ($p<0.05$).

Conclusion: For all combinations of one-step self-etch adhesives and SSA-Bond resin coating, silorane-based low shrinking composite exhibited inferior marginal adaptation than did the methacrylate-based composite.

4.2 Introduction

Even if composite materials have greatly improved since their introduction to the dental market, polymerization shrinkage still remains a major problem associated with direct resin composite restorations [1]. Bonded to the cavity walls, polymerization shrinkage creates contraction stresses [2.3] in the resin composite restoration and internal stress and deformation in the surrounding tooth structure [4] that might result in postoperative sensitivity, microleakage, marginal discoloration and gap formation [5].

Filtek Silorane (3M ESPE, St. Paul, MN, USA) is a low-shrinking composite based on non-methacrylate chemistry. The term Silorane derives from its chemical building; blocks of siloxanes and oxiranes [6]. Cyclosiloxanes are responsible for the high hydrophobicity of the material while the cycloaliphatic oxiranes guarantee reactivity. In contrast to methacrylates which polymerize through radical reaction by opening their double bonds, siloranes polymerize through cationic ring opening reaction, which is accompanied by a volumetric expansion thus reducing polymerization shrinkage to 1% [6]. This special silorane chemistry has been reported to be incompatible with methacrylate adhesive systems [6,7]. Using Filtek Silorane as restorative material requires the application of a dedicated two step self etch adhesive system - Silorane System Adhesive (SSA; 3M ESPE, St. Paul, MN, USA). The adhesive system differs from classical two-step self-etch adhesives. SSA-Primer is rather hydrophilic mild (pH 2.7) one-step self-etch adhesive that ensures bonding to the dental tissues [8]. On the contrary SSA-Bond is quite hydrophobic and bridges the hydrophilic tooth substrate with the hydrophobic silorane composite [8], preventing the contact of Silorane with water, which inhibits the cationic polymerization reaction [9]. For this reason both components, SSA-Primer and SSA-Bond, have to be polymerized separately [6].

Lower polymerization shrinkage is supposed to be associated with better bonding and improved marginal adaptation [1]. However, literature seems to be controversial about the confirmation of the positive effect of silorane low polymerization shrinkage on marginal adaptation. Several *in-vitro* studies have shown better marginal adaptation in different cavity types using silorane based composite in comparison with methacrylate [10-14] or ormocer-based [10,14] composites. On the contrary some recently published *in-vivo* studies did not find any advantage of the silorane-based composite over the methacrylate-based composite in Class I [15], Class II [16] and Class V [17] cavities after three years or ormocer-based composite in Class II cavities [18] after five years of observation. As the SSA is dedicated only to use with Filtek Silorane, the aforementioned studies are comparing marginal adaptation of low shrinking Filtek Silorane using SSA with methacrylate or ormocer resin composites in combination with their associated bonding agents. However, the use of adhesives of different bonding qualities might influence the quality of marginal adaptation as well [19,20].

Duarte *et al.* [21] have demonstrated acceptable bond strength values between hydrophobic SSA-bond and conventional total-etch methacrylate adhesive while Van Ende *et al.* [22] reported higher μTBS by using SSA in combination with methacrylate composite in comparison to silorane. In a recently published study [23] μTBS values of Filtek Silorane were not influenced when SSA-Primer was replaced by different methacrylate-based adhesive systems. These findings make the silorane composite fully compatible with methacrylate adhesives if SSA-Bond is used as an intermediate layer.

SSA-Primer contains HEMA (2-hydroxyethyl methacrylate) which is the most common hydrophilic methacrylate primer monomer incorporated in dental adhesives. It improves dentin bond strength due to its wetting enhancement effect and ensures the diffusion of co-monomers by expanding the demineralised collagen [24]. However, high HEMA concentrations promote water uptake and afterwards gradual hydrolytic degradation of the polymers, swelling and staining [25]. HEMA-free adhesives were introduced as less hydrophilic adhesives. They may reduce water sorption, increase stability of the interfacial bond and improve bonding durability [26]. Recently three *in vivo* published studies demonstrated a 5- [27,28] and 6- [29] year-long clinically acceptable performance of the HEMA-free one-step self-etch adhesive.

Studies on marginal adaptation have reported different results of methacrylate resin composites using various one-step self-etch adhesive systems [19,20,30]. So it was hypothesized in this study that different one-step self-etch adhesives might influence marginal adaptation of a silorane - based low shrinkage resin composite. Therefore, the aim of this study has been to evaluate the marginal integrity of a low-shrinking silorane (Filtek Silorane) and a “classic” microhybrid methacrylate-based (Clearfil AP-X) resin composite in Class V cavities using SSA-Primer, Clearfil S^3 bond and G-Bond one-step self-etch adhesives in combination with SSA-Bond hydrophobic resin coating.

The null hypotheses tested were that:

1) There are no differences in marginal adaptation (before/after TML) between low-shrinking silorane-based composite and standard microhybrid methacrylate-based composite.
2) Different adhesive systems have no influence on the marginal adaptation (before/after TML) of both low-shrinking silorane and standard microhybrid methacrylate-based composite.

4.3 Materials and methods

Sixty human caries-free lower third molars with completed formation were collected for the experiment. After their cleaning, the apices were sealed using an adhesive system (Optibond FL, Kerr, Orange, CA, USA) and the teeth were mounted in the centre of custom-made specimen holders using a cold-polymerizing resin (Technovit 4071, Heraeus Kulzer GmbH, Wehrheim, Germany).

To simulate dentinal fluid flow, a cylindrical hole was drilled into the pulpal chamber approximately in the coronal 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, Ivoclar Vivadent 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, by using a three-way valve, the pulp chambers were evacuated with a vacuum pump and subsequently bubble-free filled with the above solution. As of this moment, the intrapulpal pressure was maintained at 25 mm Hg throughout the testing, i.e. during cavity preparation, restoration placement, finishing and stressing. [31]

V-shaped Class V cavities were prepared (5.0 ± 0.5 mm in width, 3.0 ± 0.5 mm in height and 1.5 mm in depth) with half of the margins located in enamel and half in dentin. The cavities were finished and all the enamel margins were bevelled using 40 μm finishing diamond burs. Each bur was replaced with a new one after four cavity preparations.

The teeth were randomly assigned to six experimental groups (n=10): Group A (SSA-Primer + SSA-Bond, Filtek Silorane); Group B (SSA-Primer + SSA-Bond, Clearfil AP-X); Group C (Clearfil S^3 Bond + SSA-Bond, Filtek Silorane); Group D (Clearfil S^3 Bond + SSA-Bond, Clearfil AP-X); Group E (G-Bond + SSA-Bond, Filtek Silorane) and Group F (G-Bond + SSA-Bond, Clearfil AP-X). All the materials used in this study are described in Table 4.1.

The Silorane System Adhesive was applied to the cavities (control – Group A and B) according to the manufacturer's instructions. Application of both SSA-Primer and SSA-Bond was always followed by polymerization for 10s using L.E. Demetron II (Serial No: 792026758, Kerr, Orange, CA, USA) with a power density of 1200 mW/cm^2. In the other groups the application of SSA – Primer was replaced by Clearfil S^3 bond (Groups C and D) and G-Bond (Groups E and F) using the same polymerization time protocol.

The cavities were restored either with Filtek Silorane composite or Clearfil AP-X using a two horizontal layer technique (the first layer being placed horizontally in the cervical part up to one-half of the cavity and the second layer also horizontally in the occlusal part filling the other half of the cavity) with a polymerization protocol 20 s per layer (L.E. Demetron II). Immediately after polymerization, the restorations were finished by using fine diamond burs (Intensiv SA, Montagnola, Switzerland) and polished by using flexible aluminum oxide discs (Sof-Lex Pop-On, 3M ESPE, St. Paul, MN, USA).

After storage for one week in water at 37°C in the dark, the restored teeth were loaded in a computer-controlled chewing machine. Thermal and mechanical loading was applied simultaneously [31]. Thermal cycling was performed in flushing water changing temperatures 3,000x from 5 to 50ºC, the mechanical loading performed using $1.2 \cdot 10^6$ load cycles transferred to the center of the occlusal surface at a frequency of 1.7 Hz. A maximal load of 49 N was applied on the occlusal surface by using a natural lingual cusp taken from extracted human molars (Fig. 4.1).

Replicas of each restoration before and after loading were prepared by using a polyvinylsiloxane material (President light body, Coltene, Altstätten, Switzerland). Gold-coated epoxy replicas were prepared for the computer assisted quantitative margin analysis in a scanning electron microscope (SEM; XL20, Philips, Eindhoven, the Netherlands) and evaluated at 200x magnification. The marginal quality was expressed as percentages of continuous margins for "enamel margins", "dentin margins" and "total marginal length" [19,31].

Table 4.1: List of the materials used in the study.

Material	Composition	Application
Silorane System Adhesive – Primer LOT 7AA 3M ESPE, St. Paul, MN, USA	15–25% 2-hydroxyethyl methacrylate (HEMA); 15–25% bisphenol A diglycidyl ether dimethacrylate (BIS-GMA); 10–15% water; 10–15% ethanol; 5–15% phosphoric acid–methacryloxy–hexylesters; 8–12% silane treated silica; 5–10% 1,6-hexanediol dimethacrylate; <5% copolymer of acrylic and itaconic acid; <5% (dimethylamino) ethyl methacrylate; <3% dl-camphorquinone; <3% phosphine oxide	(1) Shake the bottle briefly before dosing so that the primer becomes less viscous. (2) Place one drop of primer into the dosing well, then close the dosing well to protect the primer from light and prevent the evaporation of the solvent. (3) Apply the primer to the entire surface of the cavity and massage over the entire area for 15 s. (4) Use a gentle stream of air until the primer is spread to an even film and does not move any longer. (5) Cure the primer for 10 s.
Silorane System Adhesive – Bond LOT 7AA 3M ESPE, St. Paul, MN, USA	70–80% substituted dimethacrylate; 5–10% silane treated silica; 5–10% triethylene glycol dimethacrylate (TEGDMA); <5% phosphoric acid–methacryloxy–hexylesters; <3% dl-camphorquinone; <3% 1,6-hexanediol dimethacrylate	(1) Shake bottle briefly before dosing so that the bond becomes less viscous. (2) Place one drop of bond in the dosing well and close the dosing well to protect the bond from light. (3) Apply the bond to the entire area of the cavity. (4) Use a gentle stream of air until the bond is spread to an even film and does not move any longer. (5) Cure the bond for 10 s.

Table 4.1 continued: List of the materials used in the study

Material	Composition	Application
Clearfil S^3 Bond LOT 041183 Kuraray Medical, Okayama, Japan	HEMA Ethanol Bis-GMA MDP Silanated colloidal silica Hydrophobic Dimethacrylate Photoinitiators Water	(1) Apply BOND to the entire cavity wall with the applicator brush. (2) Leave it in place for 10 s. (3) Use caution not to allow saliva or exudate to contact the treated surfaces. (4) Dry the entire cavity wall sufficiently by blowing mild air for more than 5 s until the bond does not move. (5) Use a vacuum aspirator to prevent the bond liquid from scattering. (6) Light-cure BOND for 10 s
G-Bond LOT 1001041 GC, Tokyo, Japan	UDMA 4-MET Phosphoric ester monomer TEGDMA Acetone Distilled Water Silica fine powder Initiators	(1) Apply to dried cavity and leave undisturbed for 10 s. (2) Dry thoroughly under maximum air pressure for 5 s in the presence of vacuum suction. (3) Light cure for 10 s
Filtek Silorane - A2 3M ESPE, St. Paul, MN, USA LOT 7AJ	5–15% 3,4-epoxycyclohexyl-ethylcyclopolymethylsiloxane 5–15% bis-3,4 epoxycyclo-hexylethylphenylmethylsilane 50–70% silanized quartz 10–20% yttriumfluoride Camphorquinone	(1) The thickness of the individual increments must not exceed 2.5 mm. (2) Cure the filling material for 20 s.
Clearfil AP-X - A2 LOT 00373A Kuraray Medical, Okayama Japan	BIS-GMA<12% TEGDMA <5% Silanated barium glass Silanated silica filler Silanated colloidal silica Catalysts Camphorquinone	(1) The thickness of the individual increments must not exceed 2 mm. (2) Cure the filling material for 20 s.

Note: The brand name Filtek Silorane is used in other countries as Filtek LS and Filtek 90

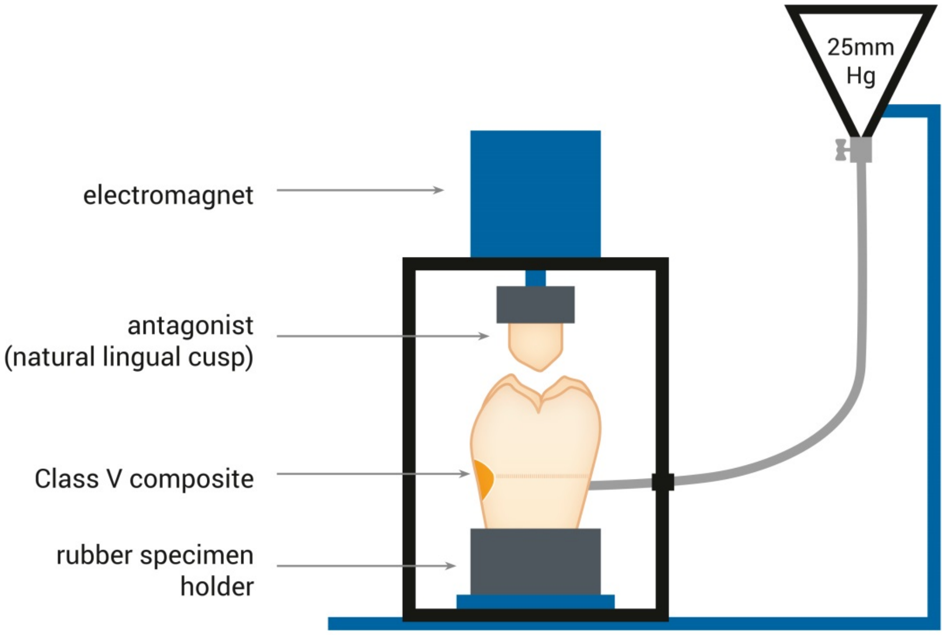

Fig. 4.1: Schematic drawing of the custom made device for occlusal loading and pulpal pressure simulation.

Statistics

Statistical analysis was performed with SPSS V21 for Mac. The dependent variables were: percentages of continuous margins *(%CM) at the total margin length*, *%CM at enamel* and *%CM at dentin*. The independent variables were *one-step self-etch adhesive system* (Silorane System Adhesive Primer/Clearfil S^3 Bond/G-Bond), *composite type* (methacrylate/silorane) and *loading interval* (before/after). To have a general overview about the performance of the different combination of composite and adhesive systems on the entire restoration margins (Total Margin Length), the 6 groups were firstly compared with 1-way ANOVA and Bonferroni post-hoc test. Then the effect of *adhesive system*, *composite type* and *loading interval* on the 3 dependent variables (total margin length, enamel and dentin margins) was evaluated with ANOVA, Bonferroni post-hoc test (for the variable « adhesive system ») and independent t-test (for the variables « composite type » and « loading interval »). The level of confidence was set to 95%.

4.4 Results

The results of marginal adaptation i.e. percentages of continuous margins (%CM) on enamel, dentin and at the total margin length, before and after loading for the 6 groups, are presented in Table 4.2.

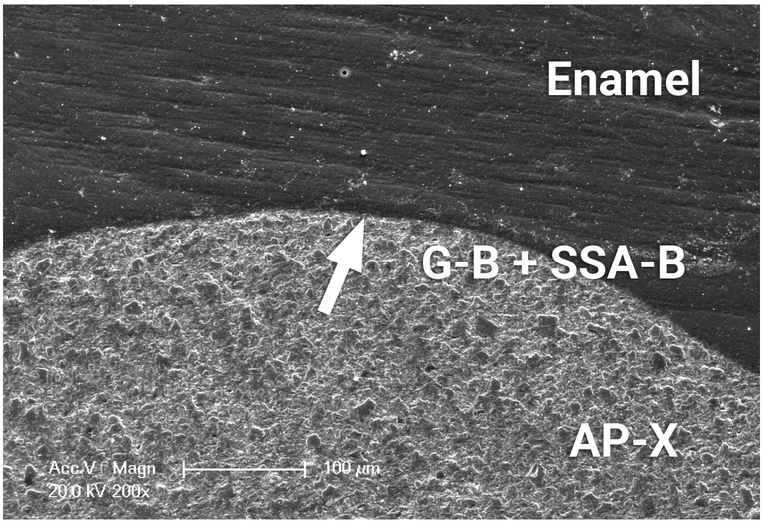

Fig. 4.2: Representative SEM micrograph of continuous margin (arrow) at the enamel-resin interface (AP-X = Clearfil AP-X; G-B + SSA-B = G-Bond + SSA-bond)

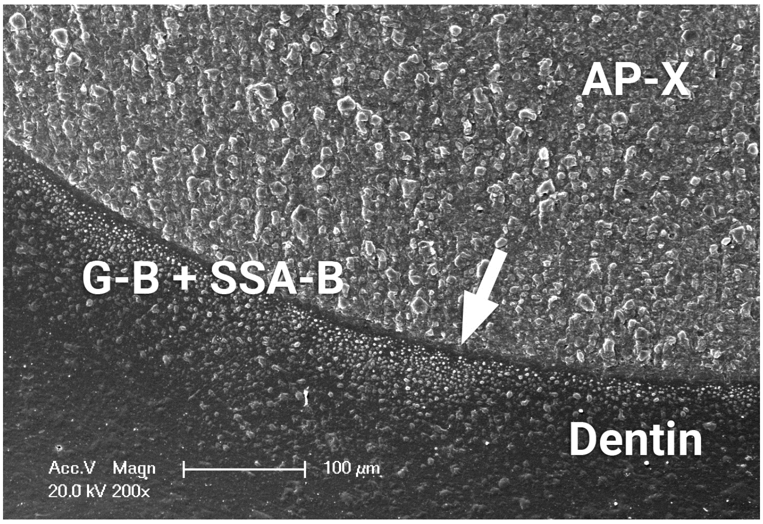

Fig. 4.3: Representative SEM micrograph of continuous margin (arrow) at the dentin-resin interface (AP-X = Clearfil AP-X; G-B + SSA-B = G-Bond + SSA-bond

Marginal adaptation was assessed by SEM for the total marginal length (i.e. enamel and dentin margins combined), and for enamel margins and dentinal margins separately. Representative micrographs are presented in Figures 4.2-4.5.

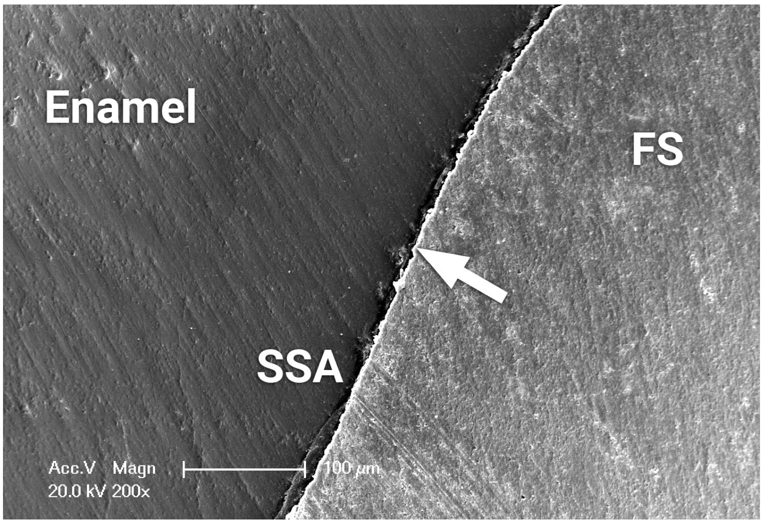

Fig. 4.4: Representative SEM micrograph of non-continuous margin (arrow) at the enamel-resin interface (FS = Filtek Silorane; SSA = Silorane System Adhesive)

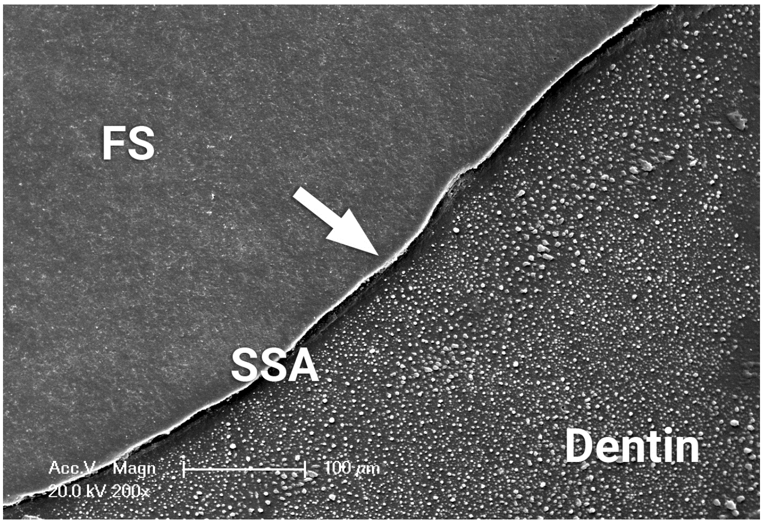

Fig. 4.5: Representative SEM micrograph of non-continuous margin (arrow) at the dentin-resin interface (FS = Filtek Silorane; SSA = Silorane System Adhesive)

At the total marginal length, the highest scores of %CM were observed in the group F (G-Bond + SSA-Bond, Clearfil AP-X: before loading (96.4 ± 3.2%) / after loading (90.8 ± 7.0%)) and the lowest scores in the group A (SSA-Primer + SSA-Bond; Filtek Silorane: before loading (73.1 ± 18.7%) / after loading (47.4 ± 20.7%)). The groups restored with methacrylate-based composite (Clearfil AP-X) performed significantly better than the silorane-based composite (Filtek Silorane) after TML ($p<0.05$). Regarding the adhesive system, for both methacrylate-based composite (Clearfil AP-X) and the silorane-based composite (Filtek Silorane), G-Bond performed significantly better ($p<0.05$) than SSA-Primer and Clearfil S^3 Bond adhesive systems. No significant differences were observed between these last two adhesive systems.

A different behavior of the materials was observed when bonded to enamel or dentin. On enamel, the variable *adhesive type* had a significant effect on the results ($p<0.05$) and the highest scores of marginal adaptation could be observed in the groups restored with the adhesive system G-Bond. The other two variables: *composite type* and *loading interval* did not have any significant effect on the results ($p>0.05$).

On dentin, the variables *composite type* and *loading interval* affected significantly the results ($p<0.05$) and their interaction was also significant, indicating that the differences that were observed between the groups at the loading intervals were due to the effect of the resin composite brand, i.e. methacrylate or silorane. The effect of the variable *adhesive type* ($p>0.05$) had no significant effect on the results. Meaning that, the highest scores of marginal adaptation were observed in the groups restored with the methacrylate based composite (Clearfil AP-X), independently of the adhesive system (Gr. B, D and E, letter A).

Table 4.2: Results of marginal adaptation: Mean (SD) of % continuous margin

Adhesive system	Composite	Before loading		After loading	
		enamel margins			
A: SSA-Primer + SSABond	FS	A B	**76.9** (17.6)	C	**43.6** (22.8)
B: SSA-Primer + SSA-Bond	APX	A B	**81.7** (14.9)	B C	**58.2** (22.2)
C: Clearfil S^3 + SSA-Bond	FS	B	**75.1** (20.8)	C	**52.4** (28.1)
D: Clearfil S^3 + SSA-Bond	APX	B	**75.2** (23.0)	B C	**59.8** (28.3)
E: G-bond + SSA-Bond	FS	A B	**91.9** (4.6)	A	**78.6** (8.1)
F: G-bond + SSA-Bond	APX	A	**93.5** (5.7)	A	**86.0** (9.4)
		dentin margins			
A: SSA-Primer + SSABond	FS	C	**68.4** (23.9)	B	**51.5** (24.8)
B: SSA-Primer + SSA-Bond	APX	A B	**95.5** (4.2)	A	**93.2** (6.8)
C: Clearfil S^3 + SSA-Bond	FS	B C	**76.5** (22.9)	B	**41.8** (33.0)
D: Clearfil S^3 + SSA-Bond	APX	A B C	**88.3** (18.7)	A	**82.8** (27.2)
E: G-bond + SSA-Bond	FS	B C	**77.4** (23.5)	B	**54.2** (31.3)
F: G-bond + SSA-Bond	APX	A	**99.6** (0.5)	A	**96.5** (5.0)
		total marginal length			
A: SSA-Primer + SSABond	FS	C	**73.1** (18.7)	C	**47.4** (20.7)
B: SSA-Primer + SSA-Bond	APX	A B	**88.1** (9.0)	B	**72.4** (16.5)
C: Clearfil S^3 + SSA-Bond	FS	B C	**75.7** (14.8)	C	**47.9** (18.7)
D: Clearfil S^3 + SSA-Bond	APX	B C	**81.0** (12.9)	B	**70.5** (18.6)
E: G-Bond + SSA-Bond	FS	A B C	**85.5** (12.4)	B	**67.3** (20.6)
F: G-Bond + SSA-Bond	APX	A	**96.4** (3.2)	A	**90.8** (7.0)

Different letters indicate significant differences between groups ($p < 0.05$)

FS = Filtek Silorane; APX = Clearfil APX

4.5 Discussion

Marginal adaptation has been described as one of the most important factors that might influence the clinical outcome of adhesive restorations [32]. Despite a recent review questioning the relevance of marginal-internal adaptation tests [33] it must be admitted that phenomena such as leakage, secondary caries, pulpal complications and fractures, which are induced by interface breakdown represent the majority of clinical failures observed in all types of direct restorations [34,35]. Then, the absence of strong correlation between clinical and in-vitro studies regarding the performance of class V restorations should not be imputed to a possible irrelevance of marginal tests but rather to methodological and sensitivity issues related to the clinical and the in-vitro studies under review. This underlines the importance of extremely well-standardized study protocols and proper simulation of the oral environment. Therefore, natural teeth, moist environment, fatigue loading comprising thermocycling and cyclic occlusal loading together with the simulation of the dentinal fluid were used in this study evaluating the marginal integrity of a methacrylate and a silorane based resin composites in Class V cavity using different combinations of adhesive systems [19,31,36].

Some one-step self-etching systems, especially those rich in HEMA, may act as semi-permeable membranes, attracting water and degrading faster than more hydrophobic adhesives [26,37-41]. To overcome this type of degradation, some investigations recommended the application of a more hydrophobic resin coating in order to provide better sealing and more durable bond strength of composite to dentin [42-44]. Actually this technique was incorporated to the original SSA protocol to bridge the difference between rather hydrophilic SSA-Primer and the quite hydrophobic silorane composite [6]. To perform the same bonding conditions SSA-Bond over polymerized one-step self-etch adhesives was applied in all groups.

The benefit of Filtek Silorane’s low polymerization shrinkage on marginal adaptation in cavities with high C-factor was proven in a previous *in-vitro* study [12] where a better marginal adaptation in Class I restorations was observed in the silorane composite resin in comparison with methacrylate composite. Nevertheless, these results may not apply to “mixed” cavities with margins located on enamel and dentin (large Class II and Class V). In these cavities the final result i.e. the percentages of “continuous margins” depends on bonding capacity of the adhesive system to two different substrates, enamel and dentin [19].

The results of our study indicate that marginal adaptation was significantly influenced by composite type, adhesive system and thermo-mechanical loading. Even before thermo-mechanical loading, none of the groups had 100% continuous margin. Our results are contradictory to previous findings in Class I cavities [12]. In Class V cavities for each adhesive system restored with both composites (Table 4.2, TOTAL MARGINAL LENGTH, groups A *vs.* B, C *vs.* D and E *vs.* F) methacrylate-based composite (Clearfil AP-X) performed significantly better than the low shrinkage silorane-based composite (Filtek Silorane). The first hypothesis was therefore rejected. When evaluating enamel and dentin margins separately a distinct behavior was observed between the silorane and methacrylate composite with respect to both tooth substrates. Marginal adaptation on enamel was not affected by the composites tested (Table 2, ENAMEL, groups A *vs.* B, C *vs.* D and E *vs.* F) on contrary dentinal margins were highly affected by the type of composite (Table 2, DENTIN, groups A vs. B, C vs. D and E vs. F).

This finding might be explained by high sensitivity of hydrophobic silorane composite – adhesive system interface to wet dentinal tissue especially in case of the simulation of dentinal fluid. Even if SSA was proved to attain an adequate dentinal seal in comparison with standard methacrylate adhesive systems [45], 100% dentinal seal is hardly obtained in normal clinical conditions. The presence of water molecules can inhibit the polymerization of silorane composite through inactivation of cationic polymerization initiators resulting in defective bonding or poorer mechanical properties [9].

According to Santini *et al.* [46] an intermediate zone of approximately 1μm between SSA-Primer and SSA-Bond was detected using micro-Raman spectroscopy, which might be the potentially weak link in the failure mechanism of silorane reconstruction. Our results of Clearfil AP-X on dentin – about 90% of continuous margins after loading - proved good compatibility between this methacrylate based composite and all adhesives tested. This fact leads to the hypothesis that the failure observed with the silorane composite must be localized between SSA-Bond and silorane. This is supported by recent μTBS findings showing high occurrence of adhesive failures between Silorane composite and SSA [22,47]. However, as the SSA-Primer and SSA-Bond are indistinguishable in SEM [46], this hypothesis must be further clarified in future research.

Three one-step self-etch adhesive systems (SSA-Primer, Clearfil S^3 Bond, G-Bond) in combination with SSA-Bond hydrophobic resin coating showed significantly different results of marginal adaptation (total marginal length) for both silorane and methacrylate composite. The second hypothesis was therefore also rejected. Two ultra-mild self-etch (SSA-Primer pH 2.8; Clearfil S3 Bond pH 2.7) and one mild self-etch (G-Bond pH 2.0) with similar nano-interaction bonding mechanism to dental tissues were compared in this study. Moura *et al.* [48] proved that the pH of adhesive per se does not have a predictive value for the bond strength value. The bonding durability depends more on the monomeric composition of the adhesive. In contrast to G-Bond both SSA-Primer and Clearfil S^3 Bond contain HEMA. HEMA is added to adhesives to improve wetting of the dentin surface and helps to mix hydrophobic and hydrophilic components into one solution. It also serves as a co-solvent to dissolve diverse active ingredients into water. However, HEMA has been considered vulnerable to hydrolysis because of hydrophilicity and water absorbtion [39]. G-Bond is HEMA-free but is supposed to be more prone to phase separation during drying of the solvent before polymerization [49].

A further difference among the three adhesives tested is their different functional monomer composition. While Clearfil S^3 Bond contains 10-MDP as functional monomer providing chemical interaction with hydroxyapatit (HAp), G-Bond contains 4-MET and phosphoric acid ester monomer. 10-MDP has been proven to have better chemical interaction with HAp compared to 4-MET [50] but its efficiency might be inhibited by the presence of HEMA [51]. Regarding the SSA, there is very limited information available in literature about the effect of the chemical interaction of SSA-Primer functional monomer (phosphoric acid-methacryloxy-hexylester) with dental tissues [8].

In respect to the methacrylate composite (Clearfil AP-X) all one-step self-etching adhesives tested showed better marginal adaptation on dentin compared to enamel margins, confirming the results of a previously published study [19]. G-Bond performed significantly better on enamel than SSA-Primer and Clearfil S^3 Bond adhesive systems, independently of composite used. Our results are in agreement with those of Blunck *et al.* [30] who observed significantly higher percentages of continuous margins on enamel with G-Bond in respect to Clearfil S^3 Bond.

4.6 Conclusions

The results of this study showed that the use of a low shrinking composite is not always the best choice to ensure good results of marginal adaptation. Not only low polymerization shrinkage and related low shrinkage stress, but also adhesive system ensuring sufficient adhesion to both tooth substrates, i.e. enamel and dentin, are the key factors for optimal marginal integrity of direct composite restorations. After thermo-mechanical loading the values of Filtek Silorane's total marginal length (%CM) were significantly lower than Clearfil AP-X. For both silorane- and methacrylate-based composites tested, G-Bond performed significantly better than SSA-Primer and Clearfil S^3 Bond under the experimental conditions used in this study.

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Chapter 5

Marginal integrity of a low shrinking resin composite in different cavities: effect of dentinal fluid simulation

This chapter has been submitted to Odontology: Ladislav Gregor, Giovanni Tommaso Rocca, Tissiana Bortolotto, Albert J. Feilzer, Ivo Krejci. Marginal integrity of a low shrinking resin composite in different cavities: effect of dentinal fluid simulation.

5.1 Abstract

Objectives: The aim of this *in-vitro* study, was to evaluate the influence of dentinal fluid simulation (DFS) on the marginal adaptation of low-shrinking silorane-based composite in Class I and Class V cavities before after thermo-mechanical loading (TML).

Methods: Four groups of teeth (n=10): Group A - Class I cavities without DFS, Group B – Class V cavities without DFS, Group C – Class I cavities with DFS and Group D – Class V cavities with DFS were restored with Silorane System Adhesive and Filtek Silorane resin composite. DFS using horse serum diluted at a 1:3 ratio with 0.9% NaCl under hydrostatic pressure of about 25 mm Hg was maintained during cavity preparation, restoration placement, finishing, polishing and TML in groups C and D. Percentages of continuous margins were quantitatively assessed with SEM before and after TML comprising 1.2 million mechanical occlusal cycles (49 N; 1.7 Hz) and 3,000 thermal cycles (5 to 50°C) applied simultaneously)

Results: 3-way ANOVA showed significant effect of *cavity type* ($p<0.05$*), TML* ($p<0.05$) and nearly significant effect of *DFS* ($p=0.051$) on marginal adaptation. However, only the interaction between the factors *cavity type* and *dentinal fluid simulation* was significant ($p<0.05$). For Class V restorations the use of DFS adversely affected dentin. There was a significant effect of the *tooth substrate* ($p<0.05$) and of *dentinal fluid simulation* ($p<0.05$) on marginal adaptation.

Conclusion: Marginal adaptation of silorane low-shrinking silorane composite in Class I and Class V cavities was influenced by the dentinal fluid simulation. Marginal adaptation on dentin was more deteriorated by the presence of dentinal fluid simulation.

5.2 Introduction

As the polymerization shrinkage and related shrinkage stress development is considered to be major drawback in resin bonded resin composite restorations (RBCs)[1] contemporary research focuses on the development of low or even non (zero) shrinking materials, which would allow for simpler, faster and more reliable restorative procedures.

Filtek Silorane (3M ESPE, St. Paul, MN, USA) is a low-shrinking resin composite based on ring opening monomer system. The term Silorane derives from its chemical building blocks; siloxanes and oxiranes [2]. Cyclosiloxanes are responsible for the high hydrophobicity of the material while the cycloaliphatic oxiranes guarantee reactivity. Silorane resin reveals lower polymerization shrinkage compared to the dimethacrylates. The polymerization shrinkage of Filtek Silorane is claimed to be about 1 vol.% [2]. In contrast to methacrylates, which polymerize through radical addition reaction of their double bonds, siloranes polymerize through cationic ring opening reaction. The cationic cure starts with the initiation process of an acidic cation which opens the oxirane ring and generates a new acidic center, a carbo-cation. After the addition to an oxirane monomer, the epoxy ring is opened to form a chain, or in the case of two- or multifunctional monomers a network is formed [2,3].

The presence of water molecules can inhibit the curing reaction of silorane resin composite through inactivation of cationic polymerization initiators resulting in defective bonding or poorer mechanical properties [4]. For this reason Filtek Silorane as restorative material requires the application of a dedicated two step self etch adhesive system - Silorane System Adhesive (SSA; 3M ESPE, St. Paul, MN, USA) – that prevents watercontact with the silorane resin composite. The adhesive system differs from classical two-step self-etch adhesives. SSA-Primer is rather hydrophilic mild (pH 2.7) one-step self-etch adhesive that ensures bonding to the dental tissues [5]. On the contrary SSA-Bond is quite hydrophobic and bridges the hydrophilic tooth substrate with the hydrophobic silorane composite [2], preventing the contact of Silorane with water.

However, the application of a separate hydrophobic solvent-free adhesive layer may reduce the hydrolytic degradation and increase the longevity of resin-dentine interfaces created with simplified adhesives [6,7]. Many laboratory experiments performed under simulated clinical conditions have reported detrimental effects of the dentinal fluid to the bond strength and sealing ability of the bonding systems [8-11]. It was hypothesized in this study that DFS performed during preparation, restoration placement and thermomechanical loading might influence the quality of marginal adaptation silorane-based low shrinking resin composite.

Therefore, the purpose of this *in vitro* study has been to evaluate the influence of DFS on the marginal adaptation of silorane-based resin composite restorations in two different cavity types before and after thermo-mechanical loading. Quantitative scanning electron microscopy (SEM) analysis based on replicas was used for evaluation in this vitro set up.

The null hypotheses tested were:

1. The evaluation of marginal adaptation does not identify different perfusion conditions (teeth with DFS vs. teeth without DFS)
2. There is no difference in marginal adaptation between enamel and dentin when margins of Class V restorations are considered.

5.3 Materials and methods

Marginal adaptation

Fourty caries-free human lower molars were collected for the experiment. After their cleaning, the apices were sealed using an adhesive system (Optibond FL, Kerr, Orange, CA, USA) and the teeth were mounted in the centre of custom-made specimen holders using a cold-polymerizing resin (Technovit 4071, Heraeus Kulzer GmbH, Wehrheim, Germany).

The teeth were randomly assigned to four experimental groups (n=10). There was no dentinal fluid simulation performed on the teeth in group A and group B. The teeth in group C and group D were prepared for the dentinal fluid simulation according to the protocol described by Krejci *et al.* [12,13]. To simulate dentinal fluid flow, a cylindrical hole was drilled into the pulpal chamber approximately in the coronal 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 34cm 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, by using a three-way valve, the pulp chambers were evacuated with a vacuum pump and subsequently bubble-free filled with the above solution. As of this moment, the intrapulpal pressure was maintained at 25 mm Hg throughout the testing, i.e. during cavity preparation, restoration placement, finishing and stressing.

In the groups A and C the Class I cavities were prepared with the use of cylindrical 80 μm diamond burs and in the groups B and D the Class V with the use of flame shape 80 μm diamond burs (Intensiv SA, Grancia, Switzerland) under continuous water cooling. Each bur was replaced with a new one after four cavity preparations. The dimensions of the Class I cavities were 5.0 ± 0.5 mm (vestibulo-lingual), 7.0 ± 0.5 mm (mesio-distal) and 3.0 ± 0.5 mm in depth. Class V cavities were 5.0 ± 0.5 mm in diameter, 3 ± 0.5 mm in height and 1.5 mm in depth with half of the margins located in enamel and half in dentin. Finally all the cavities were finished and all the enamel margins were bevelled using 40 μm finishing diamond burs.

The cavities were reconstructed using self-etch Silorane adhesive system (LOT: 7 AB, 3M ESPE AG, Seefeld, Germany) applied following the manufacturers recommendations and restored with Silorane Filtek (LOT: 7 AJ, 3M ESPE, St. Paul, USA) resin composite material. Both adhesive and restorative were polymerized using a LED curing unit (L.E.Demetron II, Serial No: 792026758, Kerr, Orange, CA, USA) with an intensity of 1,000 mW/cm^2. Class I cavities were restored using three-layer technique i.e. the first layer was placed on the pulpal floor, the second on the vestibular wall and the third on the lingual wall. Class V cavities were restored using two-layer technique i.e. the first layer was placed cervically up to one half of the cavity and the second layer occlusally, filling the other half of the cavity. Each layer was polymerized for 40 seconds. After the polymerization, the restorations were finished using fine coarse diamond burs (Intensiv SA, Grancia, Switzerland) and polished with flexible aluminium oxide discs (Sof-Lex Pop-On, 3M ESPE, St. Paul, MN, USA) with decreasing grit sizes.

After storage for at least one week in water at 37°C in the dark, the restored teeth were loaded for 10 days in a computer-controlled chewing machine. Thermal and mechanical loading were applied simultaneously [12,14]. Thermal cycling was performed in flushing water with temperatures changing 3,000 x from 5 to 50ºC with a dwell time of 2 minutes each. The mechanical stress comprised in total 1.2 million load cycles transferred to the center of the occlusal surface at a frequency of 1.7 Hz. A maximal load of 49 N was applied by using a natural lingual cusp taken from an extracted human molar.

Impressions with a polyvinylsiloxane material (President light body, Coltène-Whaledent AG, Altstätten, Switzerland) were made of each restoration before and after loading. Gold-coated epoxy replicas were prepared for the computer assisted quantitative margin analysis in a scanning electron microscope (SEM) (XL20, Philips, Eindhoven, The Netherlands) and evaluated at 200x magnification. The marginal quality was expressed as percentages of "continuous margins".

Statistical analysis:

Statistical analysis was performed with SPSS 16.0 for Windows. The dependent variables tested were *marginal adaptation* and *internal adaptation*. The independent variables were *loading interval* (before and after thermo mechanical loading), *dentinal fluid simulation* (with and without), *cavity type* (Class I and Class V) and *tooth substrate* (enamel and dentin). The influence of tooth substrate was only considered for Class V restorations as margins were composed of both enamel and dentin (mixed Class V cavity). This was not the case in Class I cavities as all margins were located on enamel. The total margin length was analyzed for each tooth specimen. Normal distribution of the data enabled the use of a 3-way ANOVA to detect significant effects and interactions between dependent variables. A post hoc t-test helped to identify differences between groups. The confidence level was set to 95%.

5.4 Results

The percentages of "continuous margins" (%CM) are presented in Fig. 5.1 and Fig. 5.2. For total margin length in Class I and Class V restorations. There was a significant effect of *cavity type* ($p<0.05$), of *testing interval* ($p<0.05$) and a nearly significant effect of *dentinal fluid simulation* ($p=0.051$) on *marginal adaptation*. However, only the interaction between the factors *cavity type* and *dentinal fluid simulation* was significant ($p<0.05$). This indicated that the differences in marginal adaptation observed between Class I and Class V restorations were due to the effect of dentinal fluid simulation.

For enamel and dentin margins in Class V restorations. There was a significant effect of the *tooth substrate* ($p<0.05$) and of *dentinal fluid simulation* ($p<0.05$) on *marginal adaptation*. If no dentinal fluid simulation was used, significantly higher percentages of continuous margins were observed on dentin when compared to enamel. However, the use of dentinal fluid simulation adversely affected dentin; the percentages of continuous margins were relatively low after loading (46.7 ± 13.8%) and no significant differences were observed with enamel (32.7 ± 17.2%).

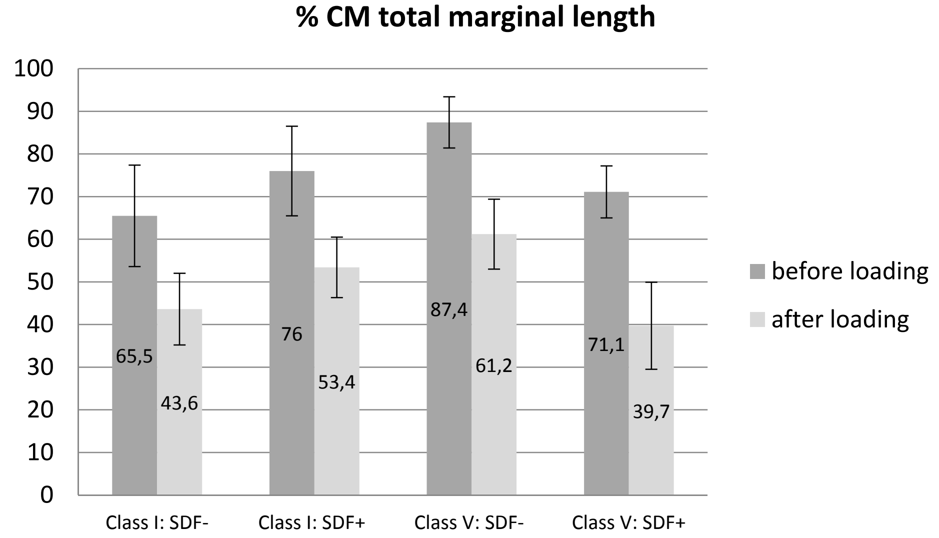

Fig. 5.1: Percentage continuous margin before and after sample loading

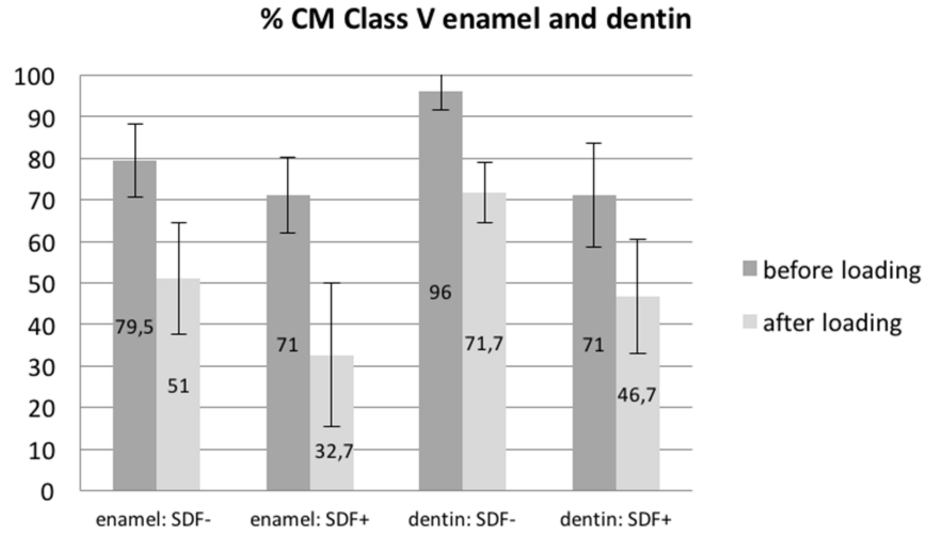

Fig. 5.2: Percentage continuous margin in a Class V cavity before and after loading

Table 5.1: Class I *vs.* Class V after TML

	No dentinal fluid simulation	**With dentinal fluid simulation**
Class I restoration	43.6 ± 8.4 **a** *A*	53.4 ± 7 **b** *A*
Class V restoration	61.2 ± 8.2 **a** *B*	39.7 ± 10.2 **b** *B*

In lower case letters (**a,b**): differences in mean values of marginal adaptation with and without dentinal fluid simulation in each group; applies to each line.
In capital letters (*A,B*): differences in mean values of marginal adaptation between Class I and Class V restorations; applies to each column.

Table 5.2: Class V after TML - enamel vs. dentin adaptation

	No dentinal fluid simulation	**With dentinal fluid simulation**
Enamel	51 ± 13.4 **a** *A*	32.7 ± 17.2 **a** *A*
Dentin	71.7 ± 7.2 **a** *B*	46.7 ± 13.8 **b** *A*

In lower case letters (a,b): differences in marginal adaptation with and without dentinal fluid simulation in each group; applies to each line.
In capital letters (A,B): differences in marginal adaptation between enamel and dentin in a Class V restoration; applies to each column.

5.5 Discussion

This study evaluated the effect of dentinal fluid simulation (DFS) on the marginal adaptation of a silorane low-shrinking resin composite in different cavities before and after loading. The chewing machine comprising thermal cycling and occlusal loading was used for this purpose.

Marginal adaptation is one of the factors of the United States Public Health Service (USPHS) criteria together with retention, staining, marginal discoloration, surface roughness and sensitivity that is used in most clinical studies to judge on the restoration's clinical success [15]. *In vitro* evaluation of marginal adaptation is based on the fact that by identifying defects at the tooth-restoration interface, an early sign of adhesive failure is already affecting the restoration before catastrophic failures like restoration loss can occur [14].

While rather simple tests were initially applied to the resin composite restorations such as dye, isotope or bacteria infiltration tests, much more sophisticated ones including eventually thermal and mechanical loading are nowadays applied [16,17]. Scanning electron microscopy (SEM) and quantitative marginal assessment proved to be complementary evaluation methods. SEM analysis provides the microscopic details of the continuity of resin-enamel and resin-dentin interface and marginal analysis allows the quantification of the rate of continuous gap-free margins on both tooth interfaces. This technique is nondestructive as by analyzing gold-coated replicas, marginal qualities can be assessed both before and after loading [12].

In clinical conditions there is an outward fluid flow across exposed dentin in response to the positive pulpal pressure [18]. Simulating physiological pulpal pressure *in vitro* has become a reliable assay to test the behavior of bonding agents, resin composites and luting agents. It expedites water penetration, polymer degradation, and droplet formation in the resin/dentin interfaces [11,19].

Hybrid layer and adhesive resin layer are permeable structures [20,21] . SSA-Primer is ultra-mild self-etch (pH 2.7) [5] adhesive system with nano-interaction bonding mechanism to dental tissues. As the smear layer is not totally removed, there is less dentinal fluid diffusion. Under this principle, a self-etching system supposed to be better than an etch-and- rinse system in cases where the intrinsic wetness is of concern [22].

As the most of dental restorations are performed under local anesthesia, dentinal fluid might be reduced when a local anaesthetic agent contain vasoconstrictor is used [23-25]. Feitosa *et al.* [26] investigated the influence of different methods of simulate pulpal pressure on bond strength and nanoleakage. In his in-vitro set-up, simulated pulpal pressure was introduced one hour after restorative procedure to simulate the length of time it takes for the effect of local anesthesia containing vasoconstrictor to wear off. However, Harnirattisa *et al.* [27] observed *in-vivo* modest effect of the local anaesthetic containing vasoconstrictor on the reduction in dentinal fluid flow through the smear–layer covered or acid etched dentin. The μTBS of self- etch and total etch adhesive to the anaesthetized and non-anaesthetized dentin were not significantly different. The seepage of pulpal fluid to the cavity after preparation might also affect the interaction of dental adhesives to dentin [28]. Thus, in the current test design, in the experimental group the DFS was employed during preparation, adhesive system application, restoration placement and thermomechanical loading.

Dentinal fluid is an ultra filtrate of pulp interstitial fluid, which is an ultra filtrate of blood plasma [29]. Different solutions such as diluted bovine [30] or horse serum [12,13,29], phosphate-buffered [31,32] or physiological saline [30,33], human plasma [33], and water [8,9] have been described for pulpal pressure simulation. The use of water for dentinal fluid simulation might overrate the bond strength of adhesives [22]. Because of possible interaction between adhesive components and plasma proteins, protein-containing liquids are preferable [22].

The results of our study indicate that marginal adaptation was significantly influenced by cavity type, loading interval and dentinal fluid simulation. Even before thermo-mechanical loading, none of the groups had 100% continuous margin. Relatively poor results of of silorane composite marginal adaptation in Class I before/after loading are in coincidence with previously published studies [34] indicating less potential for micromechanical interlocking of SSA with relatively high pH 2.7. Considering total marginal length the results (%CM) in Class V are worse than in Class I when dentinal fluid is used. On the contrary, when dentinal fluid was not used, marginal adaptation was better in Class V in respect to Class I (Table 1), probably due to a favourable C-factor and to the absence of dentinal fluid. This explains why the interaction between both factors was significant. The first hypothesis was therefore rejected.

When evaluating enamel and dentin margins separately a distinct behaviour was observed in relation to dentinal fluid simulation. The results (%CM) of dentinal adaptation were significantly better compared to enamel in absence of DFS in Class V cavity. On contrary when DFS was performed, there were no significant differences between enamel and dentin margins (Table 2). The second hypothesis was therefore partially rejected. Marginal adaptation of Filtek Silorane on dentin is highly affected by the presence of DFS. In respect to dentinal margins even before loading there were significant differences between the groups with DFS and without. In Class I cavities higher hydrolitical degradation of the SSA – dentin interface might be also possible explication of better results (%CM) after TML in teeth with DFS [35].

According to Duarte *et al.* [36] the silorane adhesive interface degradation may be explained by the presence of silane agents and siloxane groups in the adhesive composition, which are more easily hydrolytically degradable [36]. Moreover an intermediate zone of approximately 1μm localized in between SSA-Primer and SSA-Bond [37] might be the also potentially weak link to the bond instability after aging regimes, since this zone is more prone to hydrolysis and silver uptake [36].

Within the limits of this study, it may be concluded that marginal adaptation of silorane low-shrinking composite in Class I and Class V cavities is influenced by the dentinal fluid simulation. Compared to enamel, marginal adaptation on dentin was more deteriorated by the presence of dentinal fluid simulation in Class V cavity. Regarding to marginal integrity evaluation simulation of dentinal fluid should be always performed to achive more reliable clinical results.

5.5 References

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Chapter 6

Silorane, ormocer, methacrylate and compomer long-term staining susceptibility using ΔE and ΔE_{00} color-difference formulas

This chapter is published in Odontology 2015: Ladislav Gregor, Ivo Krejci, Enrico Di Bella, Albert J. Feilzer, Stefano Ardu. Silorane, ormocer, methacrylate and compomer long-term staining susceptibility using ΔE and ΔE_{00} color-difference formulas.

6.1 Abstract

Objectives: The aim of this study was to evaluate the staining susceptibility of a silorane (Filtek Silorane), an ormocer (Ceram X Duo), a methacrylate (Tetric EvoCeram) and a compomer (Dyract) exposed on the long term to various staining agents by using ΔE and ΔE_{00} color-difference formulas.

Methods: Thirty-six disc-shaped specimens were made of each of the four chemically different materials, randomly divided in 6 groups (n=6) and immersed in 5 staining solutions (red wine, juice, coke, tea and coffee) or stored dry (control) in an incubator at 37°C for 99 days. Spectrophotometric measurements by means of a spectrophotometer (Spectroshade Handy Dental, MHT) were repeated over a white (L*=92.6, a*=-1.2, b*=2.9) and black (L*=1.6 mm, a*= 1.2, b*=-1.0) background made of plasticized paper, in order to determine the color changes according to ΔE, ΔE_{00} and translucency formulas.

Results: Statistical analysis was performed by means of factorial Annova, Fisher's LSD test (post hoc) and a Spearman rank correlation between ΔE and ΔE_{00}. When analyzed over a white background mean ΔE_{00} values were highly significantly different and varied from 0.8 (Ceram X Duo/air) to 20.9 (Ceram X Duo/red wine). When analyzed over a black background mean ΔE_{00} values were highly significantly different and varied from 1.0 (Ceram X Duo and Tetric/air) to 25.2 (Ceram X Duo/red wine). Differences in translucency varied from 0.3 (Ceram X Duo/air) to 21.1 (Ceram X Duo/juice). The correlation between ΔE and ΔE_{00} over a white background was 0.9928 while over a black background it was 0.9886.

Conclusion: Significantly different staining susceptibility was found among the different restorative materials tested.

6.2 Introduction

Newly developed adhesive restorative materials, are proposed nowadays as alternatives to crowns and ceramic veneers [1-3].

Due to their resin matrix's nature, even if less hydrophilic than ancient resin composites, they are still less hydrophobic than ceramics and thus more prone to the influence of various colorants and aging [4,5]. Unfortunately, despite promising information provided by manufacturers on their potential clinical performance, there is a lack of independent scientific data regarding their behaviour in the clinical environment. In order to test their staining susceptibility an "*in vitro*" study can allow all tested materials to be submitted to standardized discoloration stress. The subsequent results can be analysed in order to understand their staining susceptibility.

The aim of this *in vitro* study was to evaluate the colour stability of different resin based restorative materials under accelerated *in vitro* test conditions. Moreover, the question whether the different calculation methods i.e. ΔE or ΔE_{00} results in different conclusions or observations was studied.

The first hypothesis tested was that the materials included into the study do not change their colour after immersion in staining agents. The second hypothesis tested was that the transparency of the materials tested does not change after immersion in staining agents. The third hypothesis tested was that no significant correlation may be found between results obtained according to ΔE and ΔE_{00} formulas. The last hypothesis tested was that ΔE and ΔE00 calculation mode leads to the same statistical conclusions.

6.3 Materials and methods

Thirty-six disc-shaped specimens measuring 10 mm in diameter were readied of each of four composite materials (Table 6.1) by gently pressing 0.2 g of the material between two glass slides (1 mm thick per 76 mm length and 26 mm wide) to the thickness of 1 mm. The composite resins were light cured for 20 seconds from a distance of 1 mm by using a Swiss Master Light curing device (serial number: M1053, EMS SA, Nyon, Switzerland) at a radiant emittance of 3000 mW/cm^2. After 24h dry storage in an incubator (INP-500, Memmert), initial colour of each specimen was assessed by a calibrated reflectance spectrophotometer (SpectroShade, Handy Dental Type 713000, Serial No. HDL0090, MHT, Arbizzano di Negar, Verona, Italy) over a white (L* = 92.6, a* =-1.2, b* = 2.9) and black (L* = 1.6, a* = 1.2, b* =-1.0) background made of plasticized paper.

Table 6.1: Materials' description

Type of material	Commercial name	Manufacturer	Color	Composition
Silorane	Filtek Silorane	3M ESPE Dental Products, St Paul, USA	A3	Dimethacrylates, Ba glass, Yttriumfluoride, mixed oxides and prepolymers
Ormocer	Ceram X Duo	Dentsply International, York, USA	E3	Methacrylate modified polysiloxane, Ba, Al Borosilicate glass
Resin Composite	Tetric EvoCeram	Ivoclar Vivadent GmbH, Schaan, Liechtenstein	A3	Bis GMA,UDMA, Ba glass, Ytterbiumfluoride, Ba-Al-Fluorosilicate glass
Compomer	Dyract	Dentsply International, York, USA	A3	UDMA, TCD resin, Alkanoyl-poly-methacrylate, Sr-F-Si glass, Sr-fluoride

Specimens were then randomly divided into 6 groups (n=6) and immersed in 2.5 ml of the 5 staining solutions tested: red wine (Côtes du Rhône (DOC), Les Arénes, Vacqueyras, France), juice (Hohes C, Eckes-Granini, Switzerland), CocaCola (CocaCola Beverages AG, Brüttisellen, Switzerland), tea (Twinings Earl Gray tea, London, England) and coffee (Arpeggio, Nespresso, Nestle, Switzerland) or kept dry (control). All specimens were stored in an incubator at 37°C for 99 days. Staining solutions were changed every two weeks to avoid bacteria or yeast contamination. After 99 days of storage, samples were removed from staining solutions, rinsed for 60s with a high pressure-hot water airbrush (0,4MPa, 135°C, Minivapor 93, Effegi Brega s.r.l., 29010 Sarmato, PC- Italy) and subsequently air dried. Spectrophotometric measurements (D65 illuminant (CIE)) were repeated to determine the colour changes and translucency changes according to the following ΔE, ΔE_{00} and TP formulas:

$$\Delta E^*_{ab} = \sqrt{(L^*_2 - L^*_1)^2 + (a^*_2 - a^*_1)^2 + (b^*_2 - b^*_1)^2}$$

$$\Delta E^*_{00} = \sqrt{\left(\frac{\Delta L'}{k_L S_L}\right)^2 + \left(\frac{\Delta C'}{k_C S_C}\right)^2 + \left(\frac{\Delta H'}{k_H S_H}\right)^2 + R_T \frac{\Delta C'}{k_C S_C}\frac{\Delta H'}{k_H S_H}}$$

$$TP = \sqrt{(L^*_B - L^*_W)^2 + (a^*_B - a^*_W)^2 + (b^*_B - b^*_W)^2}$$

where b refers to the colour coordinates over the black background and w refers to those over the white background.

Details of the methodology used in this study are explained in a precedent publication by Ardu *et al.* who already tested the behavior of resin based composite materials in an *in vitro* long term staining test [6].

Statistical analysis was performed by means of factorial Anova tests in order to test the effect of staining process (first hypothesis). Furthermore, all staining values were pooled together per composite tested and a subsequent grouping according to the Fisher's LSD test (post hoc) was applied on ΔE as well as ΔE_{00} and on the related translucency values, respectively (second tested hypothesis). Finally, a Spearman rank correlations was calculated between ΔE and ΔE_{00} (third tested hypothesis).

6.4 Results

All tested materials showed significant ($p<0.05$) colour changes after 99 days of staining immersion. When analysed over a white background mean ΔE values varied from 0.8 (Ceram X Duo/air) to 27.9 (Ceram X Duo/red wine). When analysed over a black background mean ΔE values varied from 1.0 (Ceram X Duo and Tetric/air) to 25.2 (Ceram X Duo/red wine). Differences in translucency varied from 0.1 (Tetric EvoCeram/air) to 21.1 (Ceram X Duo/juice) (Table 6.2).

Statistically significant differences among materials per staining liquid were found using a Fisher's LSD test (p-value < 0.01) for the three groups (white background, black background and transparency) and a set of classifications of the materials from A (best) to D (worst) was consequently defined (Table 6.2).

The Spearman rank correlation between ΔE and ΔE_{00} over a white background was 0.9928 while over a black background it was 0.9886, suggesting a substantial similarity between them. As a consequence, the statistical findings were the same for both calculation methods employed (ΔE or ΔE_{00}).

Table 6.2: A Fisher's LSD test for the three groups (white background, black background and transparency) and a set of classifications of the materials from A (best) to D (worst) is presented together with ΔE00 per each material and each staining liquid.

		STAINING LIQUID						
	Composite	**AIR**	**COFFEE**	**COKE**	**JUICE**	**TEA**	**RED WINE**	**ALL**
WHITE BACKGROUND	Filtek Silorane	1.9^{C}	3.4^{A}	2.4^{B}	2.3^{A}	2.9^{B}	1.8^{A}	2.4^{A}
	Ceram X Duo	0.8^{A}	23.6^{D}	2.3^{B}	10.3^{D}	10.4^{D}	27.9^{D}	12.6^{C}
	Tetric EvoCeram	1.0^{B}	16.1^{C}	1.5^{A}	3.9^{B}	8.0^{C}	21.7^{C}	8.7^{B}
	Dyract	1.2^{B}	8.9^{B}	9.2^{C}	6.0^{C}	2.0^{A}	11.0^{B}	6.4^{B}
BLACK BACKGROUND	Filtek Silorane	1.3^{B}	3.0^{A}	1.9^{A}	1.9^{A}	2.7^{A}	1.5^{A}	2.0^{A}
	Ceram X Duo	1.0^{A}	20.5^{D}	8.8^{C}	20.5^{C}	11.8^{C}	25.2^{C}	14.6^{C}
	Tetric EvoCeram	1.0^{A}	14.1^{C}	3.9^{B}	5.8^{B}	8.1^{B}	20.2^{C}	8.9^{B}
	Dyract	1.8^{B}	8.7^{B}	18.5^{D}	19.5^{D}	13.2^{C}	11.4^{B}	12.2^{C}
TRANSPARENCY	Filtek Silorane	0.8^{A}	-0.5^{A}	0.6^{A}	1.2^{A}	-0.2^{A}	-0.3^{A}	0.3^{A}
	Ceram X Duo	-0.3^{C}	-7.4^{C}	-6.4^{C}	-21.1^{C}	-6.2^{C}	-7.9^{D}	-8.2^{C}
	Tetric EvoCeram	0.1^{B}	-4.2^{B}	-2.4^{B}	-3.0^{B}	-4.0^{B}	-4.5^{B}	-3.0^{B}
	Dyract	-0.6^{C}	-12.1^{D}	-13.8^{D}	-15.7^{D}	-13.6^{D}	-5.3^{C}	-10.2^{D}

6.5 Discussion

Esthetic composite restorations are constantly exposed to staining by food and beverage colorants in the oral environment. As a result, the colour of restorations is subjected to alterations within a certain period of time. According to previous studies, the degree of colour change can be affected by numerous factors, including incomplete polymerization [7,8], water sorption [9,10], chemical reactivity [11,12], diet [13-15], oral hygiene [16,17], and surface smoothness of the restoration [18-20]. In this study we focused only on exogenous staining factors such as red wine, coffee, tea, orange juice and coke and their influence on the colour stability of the restoration materials.

All these staining agents are commonly present in today's diet and some of them have a strong potential to stain tooth coloured restorative materials and, as a consequence, they have been used in many studies [21-27]. Furthermore, some colorants [6] have showed to be able to stain more some specific resin composites maybe to their specific molecular polarity or chemical affinity. That is why we decided to use the same colorants employed in a previous study [6]. The present investigation did not include water because of previous studies that have shown that it caused no visible colour change [28]. Dry storage was used instead as the control group.

In this study we decided to test a clinically relevant condition where composite is polymerised against a Mylar strip in proximal parts of a class IV restoration. In this clinical situation the composite is richer in matrix resin [29], thus more prone to discoloration. In order to mimic this situation resin composite discs were not polished. Literature is not univocal regarding the possible influence of polishing on surface discoloration [30,31], but we think it is wiser to consider the results obtained in this study as relevant only for the above described clinical situation.

The decision of 99 days of staining immersion was made according to the estimations of Ertas *et al.* [32], who considered this period to be equivalent to 8 years of clinical ageing (24 hours *in vitro* staining corresponds to 1 month *in vivo*), thus a long term test. Generally there is little information available in the literature on the clinical correlation and only short term data are available.

The use of a spectrophotometer was made in order to avoid bias due to human eyes, to be consistent with precedent studies [6,33,34], and to allow a double evaluation over a white as well as over a black background. This double evaluation corresponds to two distinct clinical situations [35]. Black background mimics a clinical situation of Class IV composite restorations, where no tooth structure is available in the back. White background, on the other hand, is related to the situation, where one of the walls is present i.e. class I, II, II, III and veneers [6].

Considering the results obtained in this study Filtek Silorane performed better than all other materials tested. This behaviour can be related to the hydrophobicity of the resin matrix which minimizes water sorption and, consequently prevent discoloration.

Surprisingly Tetric EvoCeram and Dyract performed better than CeramX when results from all staining liquids mixed together over a white background were considered. This result is different by the one obtained when measures were taken over a black background. It can be explained by an optical analysis which revealed that Dyract change his outermost layer, becoming completely opaque and dull. Translucency parameter, in fact, witnesses the great change undergone by Dyract which, when results from all staining liquids mixed together are considered, change its opacity in an important way. This surface change can be highly disturbing in a clinical situation leading to the need of restoration replacement or at least surface modification. This behaviour can be due to the presence of HEMA and carboxylic groups which are hydrophilic thus prone to water affinity. Ceram X Duo, on the other hand seems to be highly affected by acidic juice, maybe due to the possible acidic attack of juice towards the polysiloxane components. This effect is, again, witnessed by an important change in translucency parameter.

When comparing Ceram X to Tetric EvoCeram the obtained results were in accordance with Ardu *et al.* [6], and showed a statistical superiority in favour of the latter. This can be explained by the suboptimal integration of the microfilled prepolymerized particles and the siloxane particles to the surrounding resin. These structures, in fact, even if not hydrophilic "per se" can lead to discoloration when their interface is not optimally silanized and integrated into the resin matrix thus allowing water and colorants infiltration.

Concerning the double evaluation on the basis of ΔE and ΔE_{00}, the same statistical conclusions were achieved. This is logical as the correlation between these two formulas were 0.9928 and 0.9886, respectively, when measured over a white or a black background. The double evaluation was done due to the fact that ΔE_{00} has been claimed, in the literature [14], to provide a better fit than CIELAB formula in the evaluation of colour difference thresholds having a better discrimination capacity on small colour differences than the standard ΔE. The ΔE_{00} disturbance level is reported [36], to be 2.2 while the

ΔE is 3.3 [14]. These values are reported in literature as the threshold at which 50% of observers find that the colour difference is perceived as disturbing.

Some limitations have to be considered due to the nature of the study itself: it is an *in vitro* experiment, the staining period is relatively long and corresponds to a quite heavy test for the materials tested, the surfaces of the samples were not polished and no brushing was used during the staining process. Future studies investigating all these variables are welcome for a more precise evaluation, as well as *in vivo* studies to confirm the obtained results.

6.6 Conclusions

Within the limitations of the present study, significantly different staining susceptibility was found among the different restorative materials tested.
The first three hypothesis tested were rejected. On the other hand, the fourth and last hypothesis saying that ΔE and ΔE_{00} calculation modes resulted in equal statistical conclusions was accepted.

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Summary and Conclusions

Successful long-term clinical performance of dental resin composite restorations is a wish of all practitioners. However, in spite of the fact that these materials evolved to be the first choice for replacement of lost natural tissues, several unsolved issues are still present regarding direct composite restorations. The setting of dental composites is accompanied by significant polymerization contraction, resulting, for adhesively place restorations, in the generation of stresses within the material and at the tooth-restoration interface, responsible for the most of the clinical failures. The present series of studies investigated selective properties of a new low shrinking composite.

The general introduction in **Chapter 1** provides background information on the rationale for the use of resin composites in dentistry. The classification of resin-based composites depending on their general composition is explained. Subsequently basic adhesive principles are described. The mechanism of polymerization shrinkage and related shrinkage stress with the possible clinical manifestations as well as the different options of the polymerization shrinkage reduction are provided. Finally the new low shrinking resin composite based on the silorane chemistry is introduced.

In the **Chapter 2**, the influence of volumetric shrinkage and C-factor on marginal adaptation of Class I composite restorations was investigated *in vitro*. The shrinkage kinetics were evaluated using the linear displacement (LD) and shrinkage force (SF) measurements. Different C-factors were simulated by using total or selective bonding application in Class I cavities followed and the cavities were restored either with silorane or methacrylate-based composite. Three working hypotheses tested in this article: first, that two composite materials with different chemistry would behave distinctly in terms of polymerization shrinkage, second, that the composite with lower shrinkage properties would perform better in terms of marginal adaptation before and after thermo-mechanical loading, and the third, that a low C-factor (which was simulated by avoiding adhesion to dentin) would positively influence the quality of marginal adaptation were confirmed. Silorane-based composite exhibited significantly better marginal adaptation than methacrylate-based composite.

Because of the general trend pairing new low shrinking materials with simplified adhesive systems **Chapter 3** looked at different bonding strategies of self-etch Silorane System Adhesive in relation to the marginal and internal adaptation in Class I cavities. The working hypothesis, which had to be rejected was that there is no difference in terms of marginal and internal adaptation when comparing different SSA application protocols in combination with low shrinkage silorane composite. The standard self-etch adhesive application with and without selective enamel etching was compared to selective bonding application technique. The influence of primer polymerization necessity was evaluated as well. It can be concluded, that even with low shrinkage silorane composite reaching 1vol.% shrinkage enamel pre-etching using 37% H_3PO_4 significantly improved the quality of marginal adaptation in high C-factor cavities.

In **Chapter 4** the influence of composite type and adhesive system on the quality of marginal adaptation in standardized Class V cavities before and after thermo-mechanical loading (TML) was evaluated. Two working hypothesis tested: first, that there are no differences in marginal adaptation (before/after TML) between low-shrinking silorane-based composite and standard microhybrid methacrylate-based composite, second, that different adhesive systems have no influence on the marginal adaptation (before/after TML) of both low-shrinking silorane and standard microhybrid methacrylate-based composite. Both hypothesis were rejected. Methacrylate-based composite (Clearfil AP-X) performed significantly better than the low-shrinking silorane-based composite (Filtek Silorane) in terms of marginal adaptation in Class V cavities. G-Bond performed significantly better than SSA-Primer and Clearfil S^3. The results of this study showed that the use of a low shrinking composite is not always the best choice to ensure good results of marginal adaptation. Not only low polymerization shrinkage and related low shrinkage stress, but also adhesive system ensuring sufficient adhesion to both tooth substrates, i.e. enamel and dentin, are the key factors for optimal marginal integrity of direct composite restorations.

As the silorane composite is highly hydrophobic and water might inhibit its cationic polymerization, **Chapter 5** evaluated the effect of dentinal fluid simulation on marginal integrity of silorane composite in Class I and Class V cavities. Two working hypothesis tested: first, that the evaluation of marginal adaptation does not identify different perfusion conditions (teeth with DFS vs. teeth without DFS), second, that there is no difference in marginal adaptation between enamel and dentin when margins of class V restorations are considered. The first hypothesis was rejected and the second was partially rejected. Marginal adaptation of silorane low-shrinking composite in Class I and Class V cavities is influenced by the dentinal fluid simulation. Compared to enamel, marginal adaptation on dentin was more deteriorated by the presence of dentinal fluid simulation in Class V cavity. Regarding to marginal integrity evaluation simulation of dentinal fluid should be always performed to achieve more reliable clinical results.

Chapter 6 evaluates staining susceptibility of a silorane (Filtek Silorane), an ormocer (Ceram X Duo), a methacrylate (Tetric Evo-Ceram) and a compomer (Dyract) exposed on the long term to various staining agents by using ΔE and ΔE_{00} colour-difference formulas. Thirty-six disc-shaped specimens were made of each of the four chemically different materials and immersed in five staining solutions (red wine, juice, coke, tea and coffee) or stored dry (control) in an incubator at 37°C for 99 days.

Three working hypothesis tested in the article: first, that materials included into the study do not change their colour after immersion in staining agents, second, that the transparency of the materials tested does not change after immersion in staining agents and the third, that no significant correlation may be found between results obtained according to ΔE and ΔE_{00} formulas were rejected. The last hypothesis tested, that ΔE and ΔE_{00} calculation mode leads to the same statistical conclusions was accepted. Considering the results obtained in this study, Filtek Silorane performed better than all other materials tested. This behaviour can be related to the hydrophobicity of the resin matrix which minimizes water sorption and consequently prevent discolouration. Silorane based composites are relatively new restorative materials designed specially in order to reduce volumetric contraction and contraction stress development. Compared to standard microhybrid methacrylate composites several *in vitro* studies prooved the benefit of silorane low shrinkage composite on marginal adaptation or cuspal deflection. However, from the clinical perspective, the ongoing clinical studies (up to 5 year clinical trial) didn't find the advantage of low shrinking material on the clinical performance (USPHS criteria). Although Silorane is proclaimed to reduce contraction sress development, it is not zero shrinking composite so incremental layering is still highly recomended. Not only low polymerization shrinkage and related low shrinkage stress, but also adhesive system ensuring sufficient adhesion to both tooth substrates, i.e. enamel and dentin, are the key factors for optimal marginal integrity of direct composite restorations. Enamel 37% H_3PO_4 etching improves the long term stability of Silorane System Adhesive. The silorane – enamel/dentin interface seems to be highly sensitive to wet enviroment. For this reason rubberdam application to ensure dry working field during adhesive application and silorane composite placement is recommended.

Samenvatting en Conclusies

Bij het vervaardigen van composiet restauraties is het ieders wens dat deze klinisch langdurig goed zullen voldoen. Ondanks het feit dat thans tandheelkundige composieten de eerste materiaalkeuze vormen voor het vervangen van verloren gegaan natuurlijk tandweefsel, bestaan er nog steeds onopgeloste problemen met composieten die een risico vormen om dit doel te bereiken. De verhardingsreactie van tandheelkundige composieten gaat namelijk gepaard met aanzienlijke polymerisatiekrimp, waardoor er bij aan het tandweefsel gehechte restauraties grote spanningen in het materiaal en op het tand-restauratie-hechtvlak ontstaan, die de reden kunnen vormen voor voortijdig klinisch falen. De studies die in dit proefschrift zijn beschreven bestudeerden selectieve eigenschappen van een nieuw composiet dat een minimale polymerizatiekrimp zou vertonen.

In de algemene introductie in **Hoofdstuk 1** wordt achtergrond-informatie beschreven over het gebruik van kunststof composieten in de tandheelkunde. De classificatie van kunststof composieten op basis van hun samenstelling wordt toegelicht. Vervolgens worden de fundamentele principes van hechting aan tandweefsel van kunststof restauratiematerialen beschreven. Het mechanisme van polymerisatiekrimp en krimpspanning, de aanverwante mogelijke klinische verschijnselen en de verschillende mogelijkheden van de polymerisatiekrimp te verminderen worden toegelicht. Tenslotte wordt een nieuw composiet, op basis van 'siloraan-chemie', dat een zeer lage polymerizatiekrimp vertoont geïntroduceerd.

In het **Hoofdstuk 2**, is de invloed van volumetrische krimp en C-factor op de randaansluiting van Klasse I composietrestauraties onderzocht in een *in vitro* onderzoek. De krimp kinetiek werden geëvalueerd met behulp van lineaire verplaatsing- (LD) en krimpkrachtmetingen (SF). Verschillende C-factoren werden gesimuleerd door het gebruik van volledige of selectieve hechting aan klasse I caviteiten waarna de caviteiten werden gevuld met ofwel op siloraan ofwel op methacrylaat gebaseerde composieten. Drie hypotheses werden in dit onderzoek getest: ten eerste, dat twee composiet restauratiematerialen met verschillende chemie zich duidelijk verschillend zouden gedragen wat polymerisatiekrimp betreft, ten tweede, dat het composiet met lagere krimp-eigenschappen beter zou presteren met betrekking tot de randaansluiting van de restauratie zowel voor als na thermo-mechanische belasting en ten derde, dat een lage C-factor de kwaliteit van de randaansluiting positief zou beïnvloeden. Het op siloraan gebaseerde composiet vertoonde aanzienlijk beter randaansluiting dan op methacrylaat gebaseerde composieten.

Omdat de algemene trend is dat nieuwe restauratiematerialen met lage krimp worden gecombineerd met vereenvoudigde hechtsystemen zijn in **Hoofdstuk 3** verschillende hecht-strategieën onderzocht van het zelf-etsende 'Silorane System Adhesive' (SSA) met betrekking tot de rand- en interne-aansluiting van het restauratiemateriaal in Klasse I caviteiten. De werkhypothese, die moest worden verworpen, was dat er geen verschil bestaat met betrekking tot de rand- en interne-aansluiting wanneer verschillende SSA-protocollen in combinatie met 'Silorane' composiet worden toegepast. Het standaard zelf-etsend hechtsysteem met en zonder selectieve etsing van het tandglazuur werd vergeleken met een selectieve hechting techniek. De invloed van de noodzaak om de primer eerst te polymeriseren werd ook geëvalueerd. Geconcludeerd kan worden, dat zelfs met het laag krimpende siloraan composiet (1 vol% krimp) het pre-etsen van glazuur, met behulp van 37% H_3PO_4, in aanzienlijk verbeterde kwaliteit van de randaansluiting in hoge C-factor caviteiten resulteert.

In **Hoofdstuk 4** werd de invloed van composietsoort en hechtsysteem op de kwaliteit van randaansluiting in gestandaardiseerde Klasse V caviteiten, voor en na thermo-mechanische belasting (TML), beoordeeld. Twee werkhypothesen werden getest: ten eerste dat er geen verschillen in randaansluiting (voor en na TML) tussen laag krimpend op siloraan gebaseerd composiet en standaard microhybride op methacrylaat gebaseerde composieten bestaan, ten tweede, dat verschillende hechtsystemen geen invloed hebben op de randaansluitng (voor en na TML) van zowel laag krimpend op siloraan en standaard microhybride op methacrylaat gebaseerde composieten. Beide hypotheses werd verworpen. Wat randaansluiting in de klasse V caviteiten betreft was op methacrylaat-gebaseerd composiet (Clearfil AP-X) significant beter dan het laag krimpend siloraan composiet (Filtek Silorane). Het G-Bond hechtsysteem functioneerde significant beter dan SSA-Primer en Clearfil S^3. De resultaten van dit onderzoek toonden aan dat het gebruik van een lage krimp composiet niet altijd de beste keuze is om goede resultaten met betrekking tot de randaansluiting te verkrijgen. Niet alleen lage polymerisatiekrimp en de daarmee samenhangende lage krimpspanning, maar ook het hechtsysteem dat voor voldoende hechting aan beide tandweefsels (glazuur en tandbeen) zorgdraagt, zijn belangrijke factoren voor het verkrijgen van een optimale randaansluiting van directe composietrestauraties.

Aangezien siloraan composiet zeer hydrofoob is en water de kationische polymerisatiereactie remt is in **Hoofdstuk 5** het effect van dentine-vloeistof simulatie op de randaansluiting van siloraan composiet toegepast in de Klasse

I en V caviteiten onderzocht. Twee werkhypothesen werden getest: ten eerste, dat de randaansluiting niet wordt beïnvloed door de perfusie van dentine-vloeistof (tanden met DFS *vs.* tanden zonder DFS) en ten tweede, dat er geen verschil in randaansluiting van Klasse V restauraties tussen glazuur en dentine bestaat. De eerste hypothese werd afgewezen en de tweede werd gedeeltelijk afgewezen. Randaansluiting van siloraan lage krimp composiet in Klasse I en Klasse V caviteiten wordt beïnvloed door de dentine vloeistof perfusie. Vergeleken met tandglazuur werd de randaansluiting aan dentine door de aanwezigheid van dentine vloeistof in Klasse V caviteiten meer verslechterd. Met betrekking tot onderzoek dat de randaansluiting van composietrestauraties evalueert is het toepassen van dentine vloeistof druk van belang om betrouwbare klinische resultaten te voorspellen.

Hoofdstuk 6 evalueert de gevoeligheid voor verkleuring van een op siloraan (Filtek Silorane), een op ormocer (Ceram X Duo), een op methacrylaat gebaseerd composiet (Tetric Evo-Ceram) en een compomeer (Dyract) wanneer deze langdurig worden blootgesteld aan diverse verkleurende stoffen, de door kleurverschillen met behulp van ΔE en ΔE_{00} kleurformules te bepalen. Zesendertig schijfvormige monsters werden van elk van de vier chemisch verschillende materialen vervaardigd en daarna ondergedompeld in vijf kleuringsvloeistoffen (rode wijn, fruitsap, cola, thee en koffie) of droog opgeslagen (controle) gedurende 99 dagen in een incubator bij 37°C.

Drie werkhypothesen werden getest: ten eerste, dat de testmaterialen niet van kleur zouden veranderen na onderdompeling in kleurstoffen, ten tweede, dat de transparantie van de geteste materialen niet verandert na onderdompeling in kleurstoffen en ten derde, dat er geen significante correlatie kan worden gevonden tussen de resultaten verkregen volgens ΔE en ΔE_{00} kleurformules. Alleen de laatste hypothese, dat ΔE en ΔE_{00} berekeningswijze leidt tot dezelfde statistische conclusies werd aanvaard. Filtek Silorane functioneerde in deze studie beter dan alle andere geteste materialen. Dit kan samenhangen met de hydrofobiciteit van de kunststofmatrix die waterabsorptie vermindert waardoor verkleuring wordt voorkomen.

Op siloraan gebaseerde composieten zijn relatief nieuwe restauratiematerialen, speciaal ontworpen om de ontwikkeling van volumetrische krimp en krimpspanning te verminderen. In vergelijking met standaard microhybride op methacrylaat gebaseerde composieten wijzen verschillende *in vitro* studies dat op siloraan gebaseerde lage krimp composieten wat randaansluiting en tandvervorming betreft beter functioneren. Echter, vanuit het klinisch perspectief, ondersteunen lopende klinische studies (tot 5 jaar klinisch onderzoek) niet het voordeel van lage krimp composiet wanneer de klinische prestaties (USPHS criteria) worden beschouwd. Hoewel ‘Silorane’ vermarkt wordt met de claim krimpspanningsontwikkeling te verminderen, is deze niet nul en wordt het aanbrengen van de restauratie in lagen nog steeds sterk aanbevolen. Niet alleen lage polymerisatiekrimp en de daarmee samenhangende lage krimpspanning, maar ook het hechtsysteem dat zorgt voor voldoende hechting aan glazuur en tandbeen zijn de belangrijke factoren voor het bereiken van een optimale randaansluiting van directe composietrestauraties. Glazuur met 37% H_3PO_4 etsen verbetert ook de lange termijn stabiliteit van Silorane Systeem Adhesive. De Silorane-glazuur/dentine-interface lijkt zeer gevoelig voor een vochtige omgeving te zijn. Om deze reden wordt de toepassing van rubberdam tijdens het aanbrengen van de hechtlak en het siloraan composiet om een droog werkveld te garanderen aanbevolen.

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