ORIGINAL ARTICLE

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Marginal and internal adaptation of Class II ormocer and hybrid resin composite restorations before and after load cycling

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Abstract To overcome the shortcomings of the conventional composite restorative materials, ormocer materials have been introduced over the past few years. The purpose of this study was to evaluate the marginal and internal adaptation of two ormocer restorative systems (Admira, Voco and Definite, Degussa) compared to a hybrid composite one (TPH Spectrum, Dentsply/DeTrey), before and after load cycling in Class II restorations. Standardized Class II restorations with cervical margins on enamel were divided into three groups (n=16). Teeth of each group were filled with one of the restoratives tested and its respective bonding agent. Each group was divided into two equal subgroups. The marginal and internal adaptation of the first subgroup was evaluated after 7-day water storage at room temperature and of the second after cyclic loading in a mastication simulator $(1.2 \times 10^{6} \text{ cycles}, 49 \text{ N}, 1.6 \text{ Hz})$. The occlusal and cervical marginal evaluation was conducted by videomicroscope and ranked as "excellent" and "not excellent". One thin section (150 μ m), in mesial-distal direction, of each restoration, was examined under metallographic microscope to determine the quality of internal adaptation. The occlusal and cervical adaptation of both ormocer restorative systems was similar and clearly worse compared with the hybrid composite restorative one before as well as after load cycling. Concerning internal adaptation, no gap-free ormocer restorations were detected, whereas all Spectrum restorations presented perfect adaptation. The bonding agents of the ormocers formed layers with unacceptable

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M. Chakmakchi · A. Kakaboura · C. Rahiotis Department of Operative , Faculty of Dentistry, University of Athens, Greece features (pores, fractures) whereas that of the hybrid composite achieved perfect bonding layer even after loading. The rheological characteristics of the bonding agents of the ormocer restorative systems are proposed to be responsible for their inferior marginal and internal quality in Class II restorations compared with the hybrid composite one.

Keywords Hybrid composite · Internal adaptation · Load cycling · Marginal gaps · Ormocer

Introduction

Although remarkable improvements have been conducted in the technology of the resin composite materials and the adhesive systems, clinical failures of the resin restorations are still reported, particularly when resin composites are placed in stress-bearing areas [35].

Poor marginal adaptation along the cervical margin, secondary caries and material fracture have been established as the common clinical problems of posterior resin composite restorations [13]. Furthermore, inadequate wear resistance under masticatory attrition, which leads to a loss of anatomic form, limits the application of resin composites mainly in conservative posterior restorations [29].

These problems reflect drawbacks in the resin composite materials and their adhesive systems [20]. Besides the development in the field of adhesive agents, a number of strategies have been introduced to address the shortcomings of resin composites. Introduction of new monomers has been suggested to reduce the polymerization shrinkage and to improve the physicomechanical properties of the resin composites [29].

The term "ormocer" has been used to describe a new type of resin composites which are comprised of organicinorganic hybrid copolymers. These monomers were first introduced for electronical applications [48]. Multifunctional urethane- and thioether-oligo methacrylate alkosiloxanes as sol-gel precursors have been used for preparation of the organic-inorganic copolymers. The alkoxysil groups of the silane allow the formation of an inorganic Si-O-Si network by hydrolysis and polycondensation reactions and the methacrylate groups are available for thermally or photochemically induced organic polymerization [47]. With the addition of filler particles, the ormocer composites can be used as restorative resin composite materials. In addition, specific adhesive agents have been designed to be used in combination with ormocers.

The manufacturers claim that the main advantages of the ormocers are the reduced polymerization shrinkage, the high wear resistance and the long-lasting polymermatrix stability. Although some properties of the commercially available ormocer materials have been evaluated [20, 21, 22, 42], the clinical performance of ormocer restorations has not been adequately studied and only for limited periods [19, 26, 33]. Generally, the in vivo evaluation of restorative materials and techniques, besides concerns due to ethical reasons, is very complicated, time-consuming, expensive and gives no information about the internal tissue-material interfaces. Thus, in vitro methods have been designed simulating the oral conditions in order to estimate the clinical performance and longevity of restorations. One of the advantages of the in vitro studies is their potential to provide information about the internal adaptation of the restorations, which cannot be predicted by the marginal performance [9].

Therefore, the aim of this in vitro study was to assess the marginal and internal cavity adaptation of two available commercially ormocer restorative systems and a universal hybrid resin composite one, in Class II restorations before and after load cycling. The research hypothesis was that there is no difference concerning marginal and internal adaptation between the ormocer restorative systems and the hybrid composite one under the conditions tested.

Materials and methods

The two ormocer and one hybrid restorative systems tested in this study are presented in Table 1. Forty-eight caries-free premolars

extracted for orthodontic reasons were used for the purpose of the study. Standardized Class II cavities with one proximal box and parallel buccal and lingual walls and with cervical margins located on enamel were prepared, using #331 carbide burs (Jet, Beavers, Ontario, Canada). The bur was changed after every three preparations. The cavities were made with a high-speed turbine under copious water-cooling, using a standard cavity preparation device. The dimensions of the occlusal part of the cavities were: buccal-lingual width 2 mm, occlusal depth 2 mm; and the dimensions of the proximal box were: height 4 mm, buccal-lingual width 2 mm. The enamel margins were not bevelled.

After the preparation of the cavities, the teeth were mounted into molds with the prepared proximal surface in contact with an intact molar or premolar. Afterwards, they were assigned randomly into three groups. The first group included teeth restored with Admira, the second one with Definite and the third one with TPH Spectrum combined with their respective bonding agents (Table 1). No liner or base was placed into the cavities. A transparent matrix surrounded the tooth, followed by proper wedging with wooden wedges (Hawe-Neos Dental, Gentilino, Switzerland). Each adhesive agent was applied after total etching with 37% phosphoric acid, according to the manufacturer's instructions. All the restorative materials were placed in two increments. The first one was inserted into the proximal box, 2 mm in height, and the second filled the rest of the cavity. Each layer was photopolymerized with a halogen light-curing unit (Optilux 501, Demetron, USA, output intensity 840 mW/cm², irradiation time 40 s). The restorations were finished and polished immediately after photopolymerization and removal of the matrix band, with fine-grit diamonds and Enhance polishing system (Dentsply/ DeTrey, Konstanz, Germany), so as to obtain visible outer margins. One operator prepared the cavities and placed the restorations.

Each group was divided into two subgroups (a, b). Teeth in subgroup a were stored in distilled water at room temperature $(22\pm1^{\circ}C)$ for 7 days before the evaluation and used as control. Teeth in subgroup b were immersed into a chamber of distilled water $(22\pm1^{\circ}C)$ and exposed to mastication simulator cyclic loading in a device (Willytec, Germany) described previously [17].

The protocol for cyclic loading included 1.2×10^{6} cycles, frequency 1.6 Hz, load per sample 49 N, vertical and horizontal movement 2 and 0.3 mm, respectively. The load was applied at the center of the occlusal part of the restoration. Ceramic spheres from steatite (Ceramtec, Marktredwitz, Germany) with a radius of 3 mm were used as antagonists.

The marginal adaptation of all restorations along the occlusal and cervical margins was evaluated for the loaded as well as for the specimens stored in water (control group) before and after loading and water storage, respectively, under a videomicroscope (MS-500c, Moritex, England) at ×100 magnification. The horizontal scanning frequency was 1,560 KHz while the vertical one was 50 Hz. The CCD resolution was 752×582 pixels horizontal and

Table 1 The restorative systems (material and bonding agent) tested in this study*

Restorative material (inorganic filler fraction and filler size)	Bonding agent	Batch number material/ bonding agent	Composition of bonding agents	Manufacturer
Admira (AD) 77 wt % 60.2 vol % ca. 0.7 um	Admira Bond	016725/16523	Acetone, ormocer matrix, dimethacrylate, polyfunctional methacrylate, CQ, stabilisers	Voco GmbH, Cuxhaven, Germany
Definite (DF) 77 wt % 61 vol %	Multibond	30001792/30001650	HEMA, PAMM, GPDM, ormocers matrix, Ethanol, water, CQ, fillers, BHT	Degussa AG, Hanau, Germany
TPH spectrum (SP) 77 wt % 57 vol % ca. 0.04–5 μm	Prime & Bond NT	200011106/0103001064	PENTA,UDMA, nanofillers, acetone, CQ, stabilisers, inhibitors	Dentsply DeTrey/, Konstanz, Germany

* Data were provided by the manufacturers

vertical, respectively, while the image resolution was 470 horizontal×520 vertical lines. The videomicroscope observations were conducted under standard illumination of 60 lux. The evaluation was performed according to a previously described assessment criteria [16]. The marginal adaptation per site—along occlusal and cervical, respectively—was expressed as the number of restorations ranked as "excellent" (excellent = no openings, no gaps, continuity between material-tooth) and "not excellent" (not excellent = openings, gaps, no continuity between material-tooth and tooth fracture).

For the evaluation of the internal adaptation, one thin section (150 μ m) from the middle of each restoration in a distal-mesial direction was performed, using a hard tissue microtome. The internal adaptation in terms of gap-free interface along the cervical, axial and pulpal cavity walls was examined under an optical metallographic microscope (ME 600 Eclipse, Nikon-Kogakou, Japan) at 40× magnification. The qualitative evaluation of the adhesive layer at the same cavity walls was carried out, as well. Images were recorded with parallel and crossed polarizers. Marginal and internal adaptation was graded by one examiner.

The statistical analysis evaluated the possible influence of the factor material on the marginal adaptation in occlusal and cervical area by assessing the number of excellent restorations (target variable) for subgroup a (unloaded) and subgroup b (loaded), respectively. The ordinary chi² test at a=0.05 was used to test the null hypothesis H0: the factor material does not produce different effects on the target variable concerning marginal adaptation between the ormocer restorative systems and the hybrid resin composite one, before and after load cycling. The statistical analysis was performed with the Statistical Discovery Software JMP (SAS Institute Inc. Version 4.0).

Results

Representative images from cervical margins classified as excellent and not excellent are depicted in Figs. 1 and 2, respectively.

The results from the evaluation of the marginal adaptation are summarized in Table 2. The number of restorations with "excellent" margins was statistically significantly higher for TPH Spectrum compared with Admira and Definite, before (p=0.0002) as well as after (p<0.0001) load cycling along the occlusal (p=0.0002) and cervical margins (p<0.0001). No statistically significant differences were detected between Admira and Definite at the occlusal and cervical margins before and after loading in terms of restoration ranked as excellent.

Concerning internal adaptation, a perfect one (Fig. 3) was observed in all TPH Spectrum restorations at the interfaces evaluated prior to and after loading, whereas no gap-free ormocer restorations were found under either experimental condition.

The qualitative evaluation of the bonding agent layer along the cavity walls revealed the following most remarkable features. Non-uniform layer of varied thickness (Fig. 4) or absence regionally of that layer along the cavity interfaces was observed in all ormocer restorations evaluated. In some ormocer specimens the presence of a uniform comparatively thin bonding layer either of Admira Bond or of Multibond provided perfect internal adaptation at the specific interfacial regions (Fig. 5). Inclusion of pores was observed in thick bonding layers, which were formed particularly at the internal cavity



Fig. 1 Representative image of an "excellent" restoration (no openings, no gaps, continuity between material-tooth). (Occlusal-lingual margin in a loaded Spectrum TPH restoration video-microscope 100×). *E* enamel, *RC* Resin composite



Fig. 2 Representative image of a "not excellent" restoration (not excellent = openings, gaps, no continuity between material-tooth and tooth fracture). (Occlusal-lingual margin in an unloaded Admira restoration video-microscope $100\times$). *E* enamel, *O* Ormocer, *G* Gap

angles in all ormocer restorations (Fig. 6). Furthermore, fracture lines—in six Admira and five Definite restorations—through the thick bonding layers were induced after loading (Fig. 7). These fracture lines were not observed in any of the specimens, neither in the control nor before loading. On the contrary, a continuous, uniform, comparatively thin bonding layer without pores along the

Subgroups	Occlusal margins	Cervical margins	
	Excellent	Excellent	
ADa ^a	4 (50%) ^b	2 (25%)	
ADb ^a	2 (25%)	0 (0%)	
DFa	4 (50%)	2 (25%)	
DFb	2 (25%)	2 (25%)	
SPa	8 (100%)	8 (100%)	
SPb	8 (100%)	7 (87.5%)	

 a^{a} a unloaded, b loaded

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^b Numbers in parenthesis show the percentage of restorations



Fig. 3 Image by optical microscope of a Spectrum TPH restoration $(40\times)$. A continuous, uniform, thin bonding layer is noticed. *D* Dentine, *E* enamel, *RC* Resin composite



Fig. 4 Image by optical microscope of a Definite restoration before loading (40x). Non-uniform adhesive layer along the cavity interfaces is observed. *D* Dentine, *B* Bonding agent, *O* Ormocer

cavity interfaces was detected in all TPH Spectrum restorations (Fig. 3).

Discussion

According to the results of the present study, the null hypothesis should be rejected. The factor material did produce different effects on the target variable concerning marginal adaptation before (p=0.0002) as well after load



Fig. 5 Image of a perfect adaptation of an unloaded Definite restoration achieved by a thin layer of Multibond (cervical wall, optical microscope, 40x). *D* Dentine, *E* enamel, *B* Bonding agent, *O* Ormocer



Fig. 6 Image by optical microscope of an Admira restoration before loading $(40\times)$. Inclusion of pores in thick bonding layers is noted. *D* Dentine, *B* Bonding agent, *O* Ormocer



Fig. 7 Image by optical microscope of a Definite restoration after load cycling ($40\times$). Fracture lines through the bonding layer have been formed. *D* Dentine, *B* Bonding agent, *O* Ormocer

cycling (p<0.0001). Almost the same number of "excellent" restorations was noted before as well as after loading for TPH Spectrum restorations along occlusal and cervical margins. On the other hand, a reduced number of "excellent" ormocer restorations was noted after loading with the exception of Definite at the cervical margin. Both ormocer restorative systems tested exhibited a similar marginal performance before as well after load cycling, which was statistically significantly inferior compared with hybrid resin composite system tested. The internal adaptation for both ormocer systems was also inferior compared with the one of the hybrid resin.

The load cycling protocol used in the chewing simulator is thought to represent 5 years of chewing function [18]. The chewing simulator generates impact loading in a two-body sliding test. The only difference of the system used in the present experiment with the one described in the literature [17] was that the teeth were fixed in the sample chamber, which was filled with distilled water at room temperature. A sphere with a radius of 3 mm is thought to be in the midrange of cuspal radii (2–4 mm) [15]. Steatite is a ceramic, which is considered to be relevant to the properties of enamel and proper for the two-body sliding tests [46]. Although the chewing machine generates impact loading, the conditions in the oral cavity are much more sophisticated and complicated than those this machine can produce. The period that the specimens were tested in the chewing machine in a chamber full of water cannot simulate the effect that 5 years of oral function impose upon the stability of the adhesive bond.

The clinical significance of the internal adaptation of a restoration is very clear in in vivo studies of Class V restorations where the short-term survival results of clinical function cannot predict the long-term results for some materials because of sudden retention loss of restorations [24, 39].

In the current study, the examination of the marginal and internal adaptation of the restorations by means of optical and video microscope was preferred over evaluation of replicas under scanning electron microscope (SEM). Although the two methods have been proposed to be complementary [12], the optical microscopic observation allows reliable phase identification and is less complex and time-consuming technique. It allows for the evaluation of the uniformity and continuity of the bonding agent's layer along the whole section [12]. The possible evaluation of the same specimen under various conditions and time intervals is an additional advantage of this method. A disadvantage of the optical microscope is the observation in a smaller scale than SEM. Nevertheless, for the goals of the present experiment the observation scale was adequate. On the other hand, SEM features limitations such as impression, casting and high vacuum artefacts [45].

Gap-free margins in restorations and perfect internal adaptation are considered as major determinant factors for the successful clinical performance of a restoration. The goal must be the complete marginal sealing [16, 44]. The evidence of a gap, regardless of the width and length, impairs the integrity of the restoration and endangers its longevity [34]. Even though it is proposed that water sorption and expansion re-establishes the composite resin volume, the initial adhesion is not restored and remains damaged [30]. Therefore, a ranking based on "excellent" or "not excellent" adaptation was applied as criterion in the present investigation. A multi-scale ranking system was not applied because of the relative high proportion of not rateable gaps (up to 24%) observed [38].

The hybrid resin composite system tested in this study exhibited statistically better marginal integrity along the occlusal and cervical margins of unloaded and loaded restorations than both ormocer systems did. In agreement with the findings of the current study, Class II non-stressed Definite restorations showed partly inferior marginal quality compared with some other hybrid resin restorations, using the same bonding agent but two different polymerization modes [38]. Short-term sealing efficiency of the bonding agent Prime & Bond NT was also confirmed when the excellent marginal adaptation of TPH Spectrum restorations was attained in dentin cavities after 4 weeks of water storage [49].

The marginal gap formation is a rather complicated phenomenon. It has been suggested that the marginal and internal adaptation in a resin composite restoration is influenced by the bonding ability of the adhesive agent used, the volumetric contraction of the resin composite, the stress induced during polymerization, the stiffness and the rheological properties of the resin composite [8, 32]. Generally, composite-enamel bonds survive this stress while failures are observed at composite-dentin interface [8]. Nevertheless, in the present experiment several ormocer restorations revealed insufficient sealing performance even at the occlusal enamel margin.

The inadequate sealing of a restoration permits leakage of bacteria and their products. The penetration of microorganisms into the dentin and pulp produces pulpal irritation and pathological changes [30] as well as staining in the borders of the restoration and recurrent caries. Internal gap formation may cause a loss of the restoration. The reported similar polymerization shrinkage values of the three materials tested [7, 14, 31] imply that this is not the main factor responsible for the different behaviors detected before as well after load cycling. Nevertheless, Chen et al. [5] demonstrated higher contraction stress along with a rapid contraction force build-up for Definite compared with various "packable" resin composites. This rate may be associated with the rigid matrix of Definite due to the Si-O-Si network [5]. The relatively low dynamic modulus of elasticity recorded for Admira and the low flexural modulus measured for Definite determines low ability for stress absorbance and subsequently for protection of the adhesive bond [2, 22]. However, it has been established that the inherent flexibility and elasticity of the bonding layer provides a gradient in module of elasticity between resin-bonding areas, which may absorb the stresses induced [43].

The finding of the current study that the presence in some ormocer specimens of a uniform, comparatively thin bonding layer either of Admira Bond or of Multibond ensures perfect internal adaptation at the specific interfacial regions may suggest that the discontinuous distribution of the adhesive agents accounts mostly for the low marginal quality in ormocer restorations. The rheological properties of both agents accompanied the ormocers seem to be related to the non-controlled thickness of the adhesive layers developed [10]. The non-uniform film thickness can lead to non-uniform stress distribution as well [27].

Additionally, thick bonding layers have been noticed at the internal angles of ormocer restorations, which may result in negative bonding effect [10]. The pooling of the bonding agents at the internal angles generally is likely to occur in a three-dimensional cavity preparation and represent a more relevant clinical situation [3]. The fact that the pooling of the bonding agent was not observed in any of the internal analyses for Spectrum indicate a better rheological behavior of Prime & Bond NT compared with the other two systems tested. The fact that all restorations were made from one single experienced operator excludes the possibility of handling errors. It has been demonstrated that the thicker the adhesive layer is, the higher the elastic release effect and thus, the more the stress difference is transformed in adhesive layer deformation [4]. But even if the requirement of the stress relaxation is met by increasing the thickness of an unfilled adhesive layer [6], inadequate curing and higher polymerization shrinkage may occur [10]. In addition, thick bonding layers with air porous inclusions such as the ones observed in the current study have been shown to weaken the bond and the inherent strength of the agents [25] due to the oxygen inhibition effect [10]. The thickness of the oxygen inhibition effect layer was proposed to be a function of the resin rheological properties [36], activating system and type of film former [10]. The possible stress raises function of porosities as well as the non-uniform stress distribution that they produce [27] should also not be underestimated, particularly during chewing function. Besides, fracture lines through the body of the thick layers were found after loading for the ormocer restorative systems. The latter finding indicates inadequate strength of the adhesives to withstand the forces applied during setting and chewing service of the restorations. Unlike adhesives of ormocer systems, the comparatively thin continuous layer of Prime & Bond NT may provide uniform stress distribution and presumably causes the better overall marginal and internal adaptation noticed. This thin, continuous, uniform layer seems to exhibit the appropriate properties in order to absorb the composite deformation during setting and chewing stress. The absence of air voids in Prime and Bond NT layer noticed in the current study has also been reported in a previous study [37].

Furthermore, it has been suggested that a particle-filled adhesive layer with increased toughness, such as the one of Prime & Bond NT, can facilitate a successful bonding between tooth substance and restorative material [28]. In a fractography experiment Prime & Bond NT showed strong cohesive fracture ability [40]. This may explain the absence of fracture lines through that bonding layer even after loading. Multibond is claimed also to be filled with filler particles but it seems that it is not only the presence of fillers that determinates the bonding performance of an adhesive [11].

The poor marginal quality of Definite restorations found in the current in vitro study concurs with the in vivo results obtained for that material. Definite restorations revealed a failure rate of 9.6%, mainly because of marginal problems in Class II restorations, after 1-year clinical function [26], a rate that is unacceptable according to the ADA acceptance criteria for restorative materials [1]. Manhart et al. [23] also reported that 17 out of 70 Definite direct restorations had to be replaced in a period up to 2 years. Furthermore, after 2 years of clinical function of Class I and II Definite restorations showed inferior marginal adaptation compared with that recorded at baseline [19]. It should be taken into account that the bonding agent Etch and Prime 3.0 instead of Multibond was used in all clinical trials aforementioned. On the other hand, the first clinical reports concerning the combination of TPH Spectrum and Prime & Bond NT indicate an overall acceptable behavior [41]. There is no data currently available concerning the clinical performance of Admira. Even if relatively conservative cavities were prepared for the present experiment, both ormocer restorative systems failed to achieve an acceptable marginal and internal adaptation. This presumes a worse behavior in extended restorations.

The fact that the marginal and internal adaptation of the ormocers Definite and Admira, even before the chewing simulation, was inferior to that of TPH Spectrum imposes serious questions about the durability and longevity of the bonds provided by their bonding agents.

Conclusions

The perfect marginal and internal sealing ability in hybrid resin composite system compared with the clearly inferior one of the restorations filled with both ormocers was mostly attributed, under the limitations of the present study, to their corresponding bonding agents.

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