In Vitro Study of the Structure and Adhesive Interface in Direct Restorations with Commercial Nanocomposite Materials

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The purpose of this study was: Structural characterization of two commercial nanocomposite materials (Premise ™/Kerr Corp and Tetric EvoCeram®/ Ivoclar-Vivadent); Evaluation of the marginal adaptation and adhesive interface achieved with two commercial adhesive systems (Optibond SoloPlus ™/Kerr Corp and G-Bond ™/GC). Investigation was done using electronic scanning microscopy (SEM).

Keywords: nanocomposite, adhesive system, scanning electron microscopy

By introducing nanotechnology to produce composite materials it has been possible to obtain materials with similar or better mechanical properties than previous generation of microhybrids, and with exceptional aesthetics and luster due to the particularities of nanomaterials [1-4].

Inorganic fill rates of over 80% by mass, not yet encountered in composite materials, are current to nanocomposites, improving most features and decreasing polymerization shrinkage [2,5,6]. With the nanoscale structural approach, closer to the natural size of enamel and dentin, it is expected to improve continuity and a more natural and stable interface between dental structures and these last generation restorative biomaterials [1,7].

At the same time, the improvement of adhesive techniques to enamel and dentin allowed the expansion of the area of direct composite restorations with a better prognostic over time [8].

The practitioner currently has adhesive-etching techniques (IV and V generations), self-etching adhesives (VI and VII generations) or glass ionomeric adhesives [9]. Although immediate adhesive efficiency is good for most current adhesives [10], when tested in clinical trials, there are significant variations in adhesive quality over time [11,12].

Experimental part

Eight recently extracted wisdom teeth were used. Immediately after extraction, the molars were cleaned ultrasonically and then manually on the root surface, disinfected for 3 min in 2.5% sodium hypochlorite solution, rinsed with running water and then stored in artificial saliva (Artisial® / Biocodex, France) at room temperature until use (fig. 1).

The teeth were divided into four groups of two teeth each, depending on the adhesive system and the composite material used. For ease of handling, the teeth have been fixed in addition sillicone, using universal dental impression trays.

The composition of the nanocomposite materials used is described in table 1 and that of the four adhesive systems in the table 2.

For each particular molar, two second class cavities (proximo-occlusal) with the following dimensions were prepared: vestibular-oral width 3-4 mm, depth of 2 mm, occlusal-cervical width 4-5 mm, gingival threshold placed at 1-2 mm coronary amylo-dentinal limit. Vertical cavity walls were prepared in parallel. The contours and internal angles of the cavities were rounded, with no bevel or occlusal extension cavities.

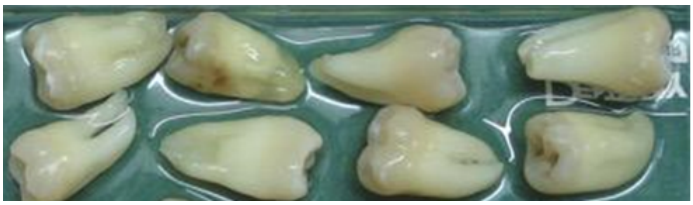

Fig. 1. Extracted wisdom teeth used in the study

Table 1
COMPOSITION AND TYPE OF NANOCOMPOSITE MATERIALS USED

Composite materials/ producer	The organic phase	Inorganic fillers
Premise ™/ Kerr Corp	Bis-fenol-A-dimethacrylate ethoxylate, TEGMA initiators, stabilizers	Prepolymerised particles, 30-50 μm, barium glass 0,4 μm, silica 0,02 μm
Tetric EvoCeram®/ Ivoclar-Vivadent	dimethacrylates	Barium glass, ytterbium trifluoride, mixed oxide

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All the authors contributed equaly to the study and the publications

Table 2

COMPOSITION AND TYPE OF ADHESIVE SYSTEM USED

Adhesive system/ producer	Composition	Class/ Stages of application
Optibond Solo Plus®/ Kerr Corp.	*Acid*: phosphoric acid 37.5% *Adhesive*: Alkyl-dimetacrylic resins, aluminoborosilicate barium glass, silicon dioxide, sodium hexafluorosilicate, ethyl alcohol *Inorganic filler*: 15% by mass	Etching-washing, two stages (generation V)
G-Bond®/GC Corp.	Monomeric esters of phosphoric acid, UDMA, 4-MET, TEGDMA, acetone, water, initiators, inorganic filler pH = 2	Self-etching adhesive, single stage (generation VII)

Table 3

DISTRIBUTION OF USE OF NANOCOMPOSITES AND ADHESIVE SYSTEMS FOR EACH GROUP

Teeth group(n=2)	Composite Material	Adhesive system
Group 1	Premise (lot nr.3047603)	Optibond SoloPlus (lot nr. 3277353)
Group 2	Premise (lot nr.3047603)	G-Bond (lot nr.0807041)
Group 3	Tetric EvoCeram (lot nr.k 32898)	Optibond SoloPlus (lot nr. 3277353)
Group 4	Tetric EvoCeram (lot nr.k 32898)	G-Bond (lot nr.0807041)

After preparation, the toilets of the cavities were made with physiological saline and restoration was carried out by applying the adhesive system corresponding to the group and then applying the composite material.

The way of combining the adhesive systems and the nanocomposite materials for each group is described in table 3.

The application of the adhesive systems was performed in a separate sequence for each of the 16 cavities being taken into operation.

Application protocol for the G-Bond adhesive system

This adhesive is part of the self-etch monocomponent adhesive systems (VII-th generation). Prior to application, the cavities were gently dried. The adhesive bottle was mixed vigorously for 5-10 s, then with an applicator it was applied by brush over the entire surface of the cavity (fig. 2). After waiting for 15-20 s, the excess adhesive was removed with a strong air spray, perpendicular to the cavity walls, for 5 s. The adhesive was then photopolymerized for 20 s with the Translux Energy® lamp (Heraeus-Kulzer)

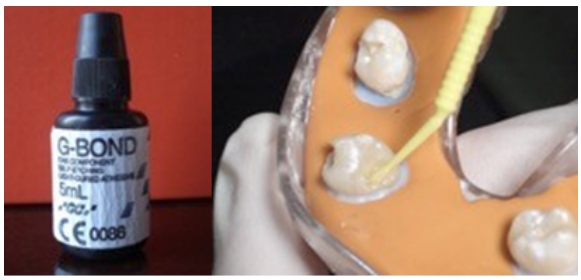

Fig. 2. Presentation and application of G-Bond single-component adhesives

Application protocol for the Optibond SoloPlus adhesive system

This adhesive used with the etching-washing technique contains the primer and the adhesive in the same vial (generation V). At the enamel surface, a 37.5% phosphoric acid gel (Gel Etchant® / Kerr Corp.) was initially applied for 15 s. Subsequently, the application continued at the dentin level for another 15 s. Then the phosphoric acid was washed for 10 s with an abundant jet of water. The cavities were dried with a moderate air spray, while maintaining the dentine wet. With the aid of an applicator, the adhesive was applied to dentine and enamel by light brushing movements for 15 s. Then the excess adhesive was removed with an air jet for 3 s then light curing was realized (20 seconds with the Translux Energy® / Heraeus-Kulzer lamp) was performed (fig. 3).

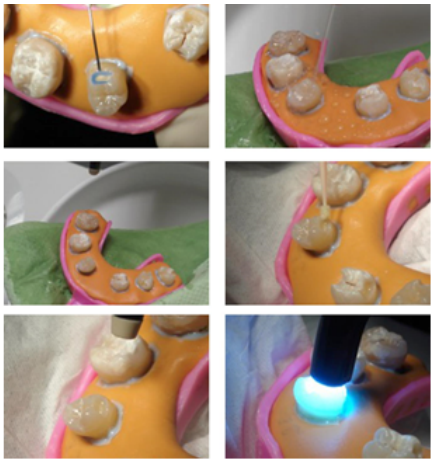

Fig. 3. Steps of application of the Optibond SoloPlus adhesive system

After application of the adhesive systems, direct restorations were achieved by the layering and polymerization of each composite material. The application of the first layer of composite material was performed in the form of a triangular prism, in the parapulpal area of the gingval wall (fig. 4). The subsequent layers were overlayed in a centrifugal direction, until the cavities were completely closed.

The thickness of the material layers was maximum 2 mm in the case of Premise and Tetric EvoCeram, and the polymerization time of 20 s/coat. The polymerization was carried out continuously with the Translux Energy® /

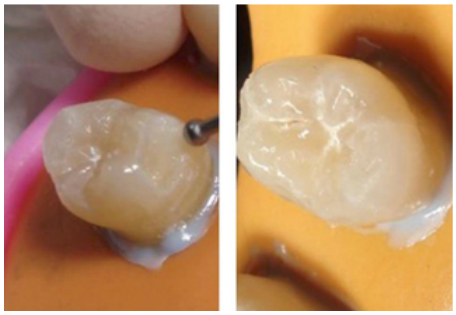

Fig. 4. Stages of layering of composite material in the cavity

Heraeus-Kulzer lamp. The direction of the beam was oblique from the direction occlusal-proximal.

After polishing, the teeth were kept in artificial saliva (Artisial® / Biocodex, France) at room temperature for seven days, then embedded in transparent autopolymer acrylic resin (Duracryl® / Spofa Dental, Kerr Corp.) and cut in a mesio-distal way by means of a diamond disc microtome (Isomet 1000 / Buehler Ltd, USA), at low speed under continuous water jet cooling (fig. 5). 1.5 mm thick specimens were obtained (fig. 6), which were cleaned in an ultrasonic tank (Emmi 20® / EMAG Technologies, Germany) in distilled water for three minutes to remove residuals generated by sectioning [13,14].

Fig. 5. Isomet 1000 Microtome

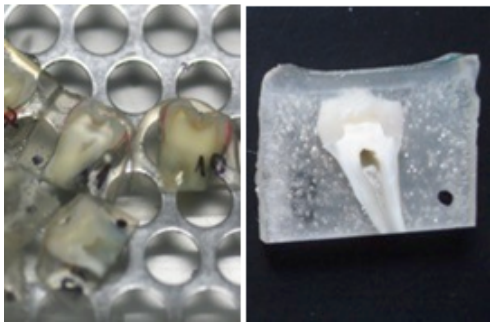

Fig. 6. Ultrasonic cleaning and section layout

Specimens were analyzed by scanning electronic microscopy (SEM) using the Hitachi S3000N (Hitachi Science Systems Ltd, Japan) electronic microscope of the University of Alicante, Spain. The images were captured in variable pressure mode, also known as *back-scattered electrons* (BSE) mode. We have evaluated: The structure of commercial composite materials; The influence of the two adhesive systems used on the adhesive interface; The integrity and continuity of the interfaces between the tooth-adhesive and the adhesive-composite material respectively.

Results and discussions

Scanning electron microscopy images have described the adhesive interface and structure characteristics for the nanocomposite materials and for studied adhesive systems. Initially, smaller magnifications (40-100x) were generally used for overall assessment of the restorations and adhesive interface, then 200-1000x magnification for composite material analysis, and 1000-1500x for the study of the hybrid layer.

The SEM images for group 1 showed a thicker, continuous and uniform adhesive layer in the structure of which micronized filler particles with good junction can be detected at both the composite material interface and the enamel and dentine (fig. 7a,b,c). The hybrid layer is homogeneous, and some infiltrated dental channel can be detected in some images (fig. 7b). The structure of the nanocomposite material Premise presented, at a magnification of 100x, a homogeneous image, with 20-50 ìm diameter, light gray, polygonal, rounded edges. Some samples showed air voids up to 100 ìm in diameter. At higher magnifications (500-1000x) the particles described were no longer detected, the image becoming homogeneous, with a high density of rounded, open-shaded, and submicron-shaped polygonal particles (fig. 7a,b,c).

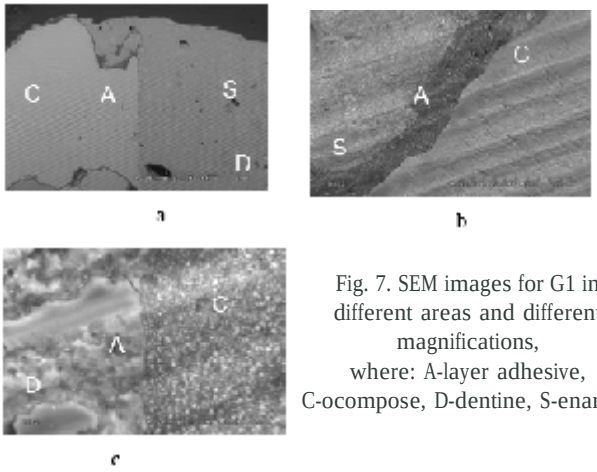

Fig. 7. SEM images for G1 in different areas and different magnifications, where: A-layer adhesive, C-ocompose, D-dentine, S-enamel.

The electron microscopy revealed in the case of the group 2 a tight, uniform adhesive layer that incorporated the smear layer from the surface of the dental plague and sends very short extensions into the dentinal canaliculi (fig. 8a,b,c). The structure of nanocomposite material was identical to that of group 1.

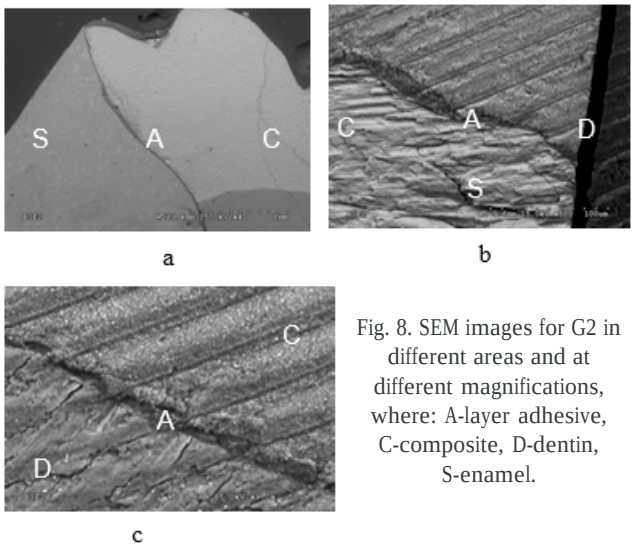

Fig. 8. SEM images for G2 in different areas and at different magnifications, where: A-layer adhesive, C-composite, D-dentin, S-enamel.

In the case of group 3, a consistent, uniform and thicker adhesive layer with sub-micron particles detectable in the structure was noted, and a continuous interface and a homogeneous hybrid layer with intracanalicular extensions (fig. 9a,b,c) was realized. The image of the Tetric EvoCeram nanocomposite material revealed two homogeneous particle types at 100x magnification: light gray, polygonal particles with rounded edges, 5-75µm in size, or very rare dark gray, round or ovoid particles with Diameter between

25-50 μm. At 1000x magnification, only rounded, light-colored particles with variable but submicron diameters were highlighted. Air inclusions similar to those described in other nanocomposites could have been more rarely observed (fig. 9a,b,c).

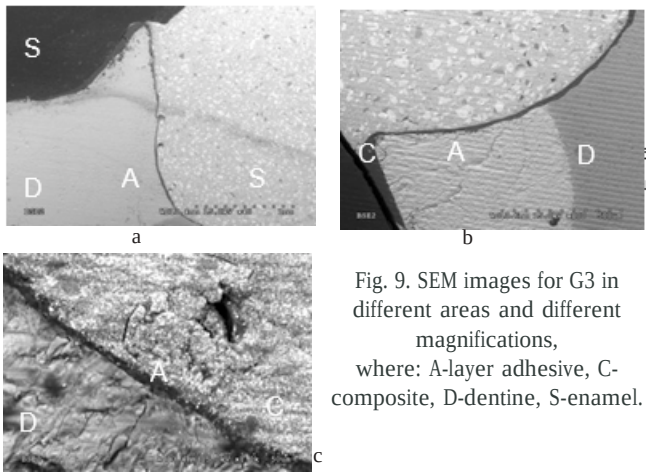

Fig. 9. SEM images for G3 in different areas and different magnifications, where: A-layer adhesive, C-composite, D-dentine, S-enamel.

Group 4 SEM images showed a uniform, thin adhesive layer with very good adaptation to both interfaces. The smear-layer was embedded but adherent extensions in the dentinal canaliculi could not be identified (fig. 10a,b,c). The structure image of nanocomposite material was similar to group 3.

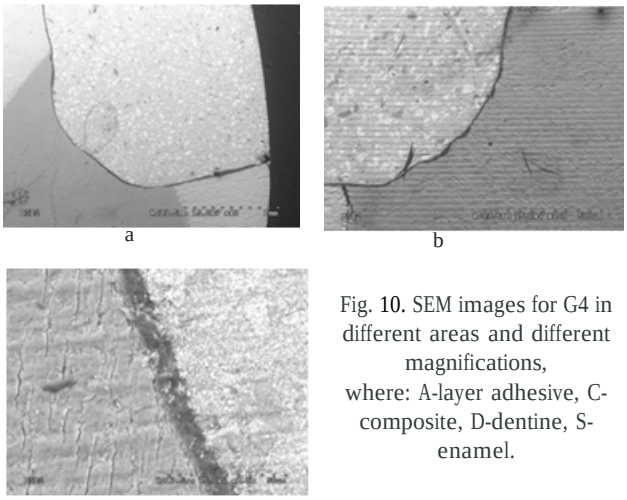

Fig. 10. SEM images for G4 in different areas and different magnifications, where: A-layer adhesive, C-composite, D-dentine, S-enamel.

The parallel striations present on all images were interpreted as sectional artifacts.

Diacrylic nanofillers composite resins have been introduced into the restorative therapeutic arsenal over the past decade in order to provide the clinician with improved mechanical properties, aesthetics and chandelier as close as possible to dental tissues, lowering the polymerization contraction by increasing the percentage of inorganic filler, Better maneuverability [1-4,6,15-17].

In this approach, the characteristics of the composition and size of the inorganic filler particles are essential: some materials - ex: Filtek Supreme™/3M Espe - contain only isolated, or aggregated nanoparticles, while most nanocomposites have a hybrid composition - And Tetric EvoCeram - combining the mechanical advantages of barium borosilicate bottles, increasing the percentage of filler by means of prepolymerized and aesthetic particles, polish and handling of nanoparticles and nanomodifiers [5,15,17].

Scanning electron microscopy images at magnification up to 100x revealed in the case of Premise nanocomposites (figs. 7a and 8a) and Tetric EvoCeram (figs. 9a,b and 10a,b) polygonal particles of grayish-gray and with a diameter of 5-75μm, which could not be observed at magnifications over 500x, which plead for their identification as prepolymerized particles according to the manufacturers specifications [15,18]. At magnifications over 1000x, both nanocomposites (figs. 7b,c, 8b,c, 9c and 10c) highlight a homogeneous structure with uniformly distributed submicron particles with rounded edges identified as the filler Barium glass [15,18,19].

The nanoparticle particles could not be highlighted at the magnifications used in this study, being observable with SEM at a magnification of 5000-10,000x [1,3,7] or by TEM at magnification over 60,000x [17].

According to the classification of Bayne et al. 2002 [20], nanofilled and nanohybrid composites (the largest particle denominates the hybrid class) contain particles with a size between 0.005-0.01μm. Nonetheless, we find a wide range of names in the RDC with commercially available names, which do not necessarily take into account this classification: Filtek ™ Supreme Universal Restorative, Nanobiber Composite (Grandio®), Universal Trimodal Composite (Premise ™), Ceramic nano-optimized model (Tetric EvoCeram®), etc. Nanocomposites have a more uniform dimensional mean of particles, while nanohybrids possess many different types of particles [2]. Characteristic of nanocomposites is the incorporation of a higher percentage of nanofibers relative to the rest of the composites due to the characteristics of the non-aggregated and silanized nanoparticles acting as a liquid and not as a solid, without adversely altering the rheological properties of the composite material [2,6].

The continuity of the interface at the adhesive-composite junction and the adhesive-dental tissue also advocates for good handling of these materials and supports a lower polymerization contraction by the increased percentage of filler [2,3,15,18]. However, most specimens in Premise, and a minority of those with Tetric EvoCeram showed air inclusions up to 100ìm in diameter; the vast majority of them were in the mass of the filling, some of them also visible at the adhesive interface (figs. 7a, 9a,b,c and 10a,b).

To improve the quality of the restoration, the cavity must be obstructed without voids and porosities. In the present study, only a small part of the sections were without porosities. Previous studies have shown that it is probably not possible to avoid the incorporation of bubbles or porosities because the curing materials taken from the syringe contain porosities in a volumetric percentage of 0.05-1.5% [21]. The quoted study showed an increase in the number of porosities with the viscosity of the material, which is in line with the present observations: the nanocomposite Tetric Evo Ceram is less viscous than Premise and has the lowest number of porosities.

Clinical relevance of the porosities is unclear: their presence in large numbers at the composite-dental tissue interface may lead to micro-infiltration and failure of restoration by secondary caries or dental pulp damages; however, such clinical failures due to porosities are rarely reported [22].

The main purpose of dental adhesives is to ensure retention for composite restorations or cemented restorations. In addition to resistance to mechanical stress, especially to those caused by the stress of polymerization contraction of the overlying composite, a good adhesive must also prevent microinfiltrations along the edges of the restoration [23-26]. Clinical failure of restorations occurs mainly through inappropriate sealing, resulting in marginal dyspromia, only by loss of retention [27,28].

A comparison of contemporary adhesives reveals that three-step ethanol-water-based adhesives through the washing-etching techniques remain the *gold standard* in terms of durability of adhesion. Any simplification in the clinical application procedure results in a decrease in adhesive efficacy. Only two-step self-etching adhesives are the closest to this standard, with the added benefit of ease of handling and less sensitive techniques [25].

Conclusions

The present study highlighted the electronomicroscopic characteristics of commercials composite materials and adhesive systems used:

The Premise and Tetric EvoCeram Nanocomposites exhibit structural homogeneity and characteristics of the inorganic filler with the prepolymerized particles and barium-borosilicate-based filler with a submicron mean size.

Visualization of nanoparticles is not possible at the SEM magnifications used in this study.

The studied nanocomposites have a variable number of porosities and air bubbles, with a difficult clinical relevance.

Adhesive systems of various generations used produce a continuous interface and good marginal adaptation in most cases.

The most homogeneous and thick hybrid layer is generated by the Optibond Solo Plus adhesive system, but the most watertight interface belongs to the specimens where the G-Bond adhesive was used.

In vivo studies are necessary to evaluate the durability of adhesion in the case of self-etch adhesives in one or two steps

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