

Methacrylate Versus Ormocer-Based Composites: Impact of Preheating on Repair Bond Strength

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Abstract:

Objective: To evaluate and compare the impact of preheating on the repair bond strength of Methacrylate and Ormocer based composite. **Materials and Methods:** A total number of 24 standardized cuboid composite blocks were fabricated with the aid of teflon split mold (25×4×4) mm dimensions. Half of these composite blocks were made from a universal nanohybrid methacrylate-based composite and the other half was made from a universal nanohybrid Ormocer based. The top surfaces of all mounted specimens were surface treated with sandblasting, silane coupling agents, and universal adhesive of the same brand of composite. Four cylindrical composite rods (2 mm height, 3 mm internal diameter) were built over each composite block and filled with the repair composite from the same material with the aid of tygon tubes. Composite blocks of both groups were divided into two sub-groups according to the previous treatment of repairing materials (preheated or not preheated). Then half of the blocks with their attached repair materials were stored for 24 h to evaluate the immediate bond strength and the other half was stored in distilled water for 6 months to evaluate the bond durability. Shear bond strength was measured using a universal testing machine (Instron), and the collected data were statistically analyzed with three-way ANOVA and post-hoc Tukey test. **Results:** The outcome of three-way ANOVA revealed that there was a statistically significant difference between the repair bond strength of Methacrylate & Ormocer groups immediately and after 6 months of storage (p=0.03). No statistically significant difference was found between the repair bond strength of the preheated and non-preheated immediately and after 6 months of storage (p=0.7). **Conclusions:** Immediate and delayed repair bond strength of methacrylate-based resin composite was superior to Ormocer based composite within the preheated group. Preheating of both methacrylate and Ormocer composite did not affect immediate and delayed repair bond strength. Ormocer based composite groups could not succeed to maintain the repair bond strength after six months of storage, while methacrylate could maintain.

Introduction:

Dental composite restorative materials are now the most popular direct restorative material for both anterior and posterior teeth due to the increasing aesthetic demands of patients.¹ Additionally, due to dental amalgam's detrimental effects on the environment, dental composites have replaced it as the preferred restorative material in the majority of nations, due to the negative environmental side effects of dental amalgam.^{2,3}

Ormocer, organically modified ceramics are composed of inorganic-organic co-polymers with inorganic silanated filler particles.⁴ Ormocers combine the advantages of the organic polymers, such as flexibility and impact resistance, and the advantages of the inorganic materials, such as thermal stability, mechanical strength, and chemical resistance.⁵ Many studies reported that Ormocer showed lower polymerization shrinkage and better biocompatibility compared to other composite resin materials.

Moreover, they are reported to have enhanced fracture and wear resistance when compared with resin-based composites.⁶ It can double the conversion of monomers and improve the physical properties of the material.⁷

A recent development in the field of dentistry is the pre-warming of resin composite restorations.⁸ Preheating raises the composite's temperature, improves radical and monomer mobility for higher overall conversion, and speeds up the polymerization process.⁹ leading to increased surface hardness, flexural and radial tensile strength.¹⁰

Different surface treatments, mechanical, physical, chemical, or a combination of all were evaluated extensively in previous studies.^{11,12} Since the old composite no longer has its oxygen inhibited layer, the use of a silane primer enhances the wetting of fresh resin composites to aged composite substrates, promotes chemical bonding of resin to filler particles and increases the flow of low-viscosity adhesives on irregular surfaces.¹³ Additionally, the application of bonding agents could be responsible for chemical bonding to the organic matrix of the repaired resin composite.¹⁴

Recently, several studies focused on the repair of methacrylate-based bulk-fill resin composites, either with conventional resin composites or with the bulk fill resin composites, either with conventional resin

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Table1: Materials used in the study

Material (Description)	Composition	(Batch#)	Manufacturer
Grandio(Universal Nano –hybrid resin restorative material)	- Bis-GMA, UDMA, TEG-DMA - <u>Filler</u> : silicium dioxide nano fillers (20-50 nm) glass ceramic micro fillers (1 µm) - <u>Filler content</u> : 87% (w/w), 71.4% (v/v).	732242	VOCO GmbH, Cuxhaven, Germany
Admira Fusion (Universal Nano – hybrid restorative material)	- Aromatic and aliphatic dimethacrylates - Methacrylate-functionalized polysiloxane - Barium-aluminum-glass, pyrogenic silicon dioxide - <u>Filler</u> : Glass ceramic (1-1.5 µm) - <u>Filler content</u> : 84% (w/w), 60% (v/v).	1905236	VOCO GmbH
Futurabond M+ (Universal adhesive)	- HEMA 10-25 %, Bis-GMA 10-25 %, Ethanol 10-25 %, Acidic adhesive monomer 2.5-5 % UDMA 2.5-5 %, Catalyst, dimethacrylates, fumed silica,CQ, BHT, water.	1428143	VOCO GmbH
Admira Bond (Ormocer-based etch-and-rinse adhesive)	- HEMA, Bis-GMA, acetone,Ormocer resin, dimethacrylates, organic acid, CQ, BHT NaF.	1421529	VOCO GmbH
Silane (Porcelain primers)	Ethanol, silane coupling agents.	1200004083	Bisco Inc. Schaumburg, IL. USA

composite properties, investigate the conversion of double bonds and the hardness of the composite.^{16,17}

However, the previous studies did not gain much attention and had little information about the repair potential of Ormocer-based composite and the effect of preheating on the repair bond strength. So, it would be prudent to investigate the impact of preheating on the repair bond strength of Methacrylate and Ormocer based composites.

Material and Methods:

Materials: Two packable dental composites (a methacrylate-based (Grandio) and an Ormocer-based (Admira Fusion) were used in this study. An Ormocer-based etch-and-rinse adhesive (Admira Bond), a universal adhesive (Futurabond M+), and Silane coupling agent (porcelain primers) were also used as intermediate agents before repair procedures. All materials were used and manipulated according to manufacturers' instructions, Table 1.

Methods :

1-Specimen preparation: A total number of 24 standardized cuboid composite blocks were fabricated using a customized Teflon mold with (25× 4 × 4) mm dimensions. Half of these composite blocks were made from a universal nanohybrid methacrylate-based composite and the other half was made from a universal nanohybrid Ormocer based composite. A transparent polyester strip was applied against a microscope glass slide. Then, the composite material was packed into the mold with a gold-plated ball burnisher applicator. The adequately Filled mold was covered by another polyester strip and glass slide.

2- Surface treatment of specimen: All the specimens were mounted in an acrylic mold.

The top surfaces of all mounted specimens were sandblasted with Al₂O₃ powder particles. Then, the Silane coupling allowed to set for 30 seconds. For the universal nanohybrid methacrylate-based composite blocks, adhesion was done by a universal adhesive; Voco Futurabond M+. For the universal nanohybrid Ormocer-based composite blocks, adhesion was done by an Ormocer adhesive (Admira bond).

3. Specimen grouping and Repair process: Four cylindrical composite rods (2 mm height, 3 mm internal diameter) were built over each composite block and filled with the repair composite from the same material with the aid of tygon tubes. Then photopolymerized for 20 s, and then removed. Composite blocks of both groups were divided into two sub-groups according to the previous treatment of repairing materials (preheated or not preheated) (n=48). Half of the blocks with their attached tygons were stored for 24 h to evaluate the immediate bond strength and the other half was stored in distilled water for 6 months to evaluate the bond durability. Thermocycling was for 5000 cycles between 5°C and 55°C. Shear bond strength was measured by Universal Testing Machine e100kn Instron Computer Universal Testing Machine (Instron). A shear force was applied using a shearing blade parallel to the adhesive interface. The load was applied to the interface at a cross-head speed of 1 mm/min until failure and the stress-strain curve was analyzed with the machine's software program. The SBS was calculated in Megapascals (MPa) by dividing the fracture load (Newton) by the repair surface area (mm²), Figure 1. Selected samples were chosen to visualize the topography of samples after failure, specimen was gold-sputtered by a 150-A° thin gold layer; the surface topography was then evaluated under a Scanning Electron Microscope JEOL, JSM-65101V)

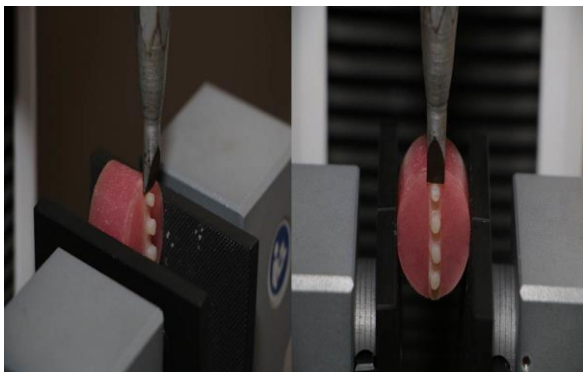


Figure 1: Fixation of mounted sample in a universal testing machine, the blade was at the interface and parallel to the longitudinal axis of samples

(at $\times 1000$ magnification. The failure modes were determined, Figure 2.

Cohesive A: failure within the old composite.

Cohesive B: failure within the repaired composite.

Adhesive: failure within the adhesive layer.

Mixed: (cohesive A with an adhesive) or (cohesive B with adhesive or (cohesive A with cohesive B).

Statistical analysis: The means Shear bond strength of the four tygon tubes from each block were averaged and the overall mean of the group was calculated from 12 blocks. The collected data were tabulated, coded, then analyzed using the computer program; Statistical package for social science (SPSS) version 26.0 (IBM, NY, USA). Data were presented as the mean, standard deviation, and percentage. The significance level was set at $p < 0.05$. Three-way ANOVA was conducted to evaluate the effect of "material", "preheating", and "storage time" and their interactions on SBS. Post-hoc Tukey multiple comparison test with a statistical significance of $p < 0.05$ was made to compare different surface treatments and storage times between the two materials.

Result:

-Within the preheated group, there is a statistically significant difference between the repair bond strength of Methacrylate & Ormocer groups immediately and after storage with P value 0.03, Table 2.

-Within the non-preheated group, there is no statistically significant difference between the repair bond strength of Methacrylate & Ormocer groups in the immediate subgroup with P value 0.7. While there is a statistically significant difference between the repair bond strength of Methacrylate & Ormocer groups in the delayed subgroup with P value 0.002, Table 2.

-Within the methacrylate group, there is a statistically significant difference between repair bond strength of preheated and non-preheated in immediate and delayed subgroups with P value. 0008, Table 2.

-There is no statistically significant difference between repair bond strength of preheated and non-preheated in immediate and delayed subgroups with P value 0.47, Table 2.

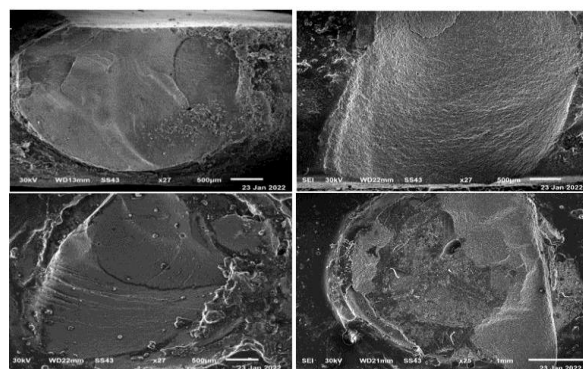


Figure 2: Representative images of failure mode showing that Methacrylate immediate heated presented cohesive A failure within old composite whereas Methacrylate immediate non-heated presented 100% cohesive failure.

-Within the methacrylate group, there is no statistically significant difference between the repair bond strength of immediate and delayed groups in preheated and non-preheated subgroups with P value 0.32, (Table 2).

There is a statistically significant difference between repair bond strength of immediate and delayed groups in preheated and non-preheated subgroups with P value < 0.001 within Ormocer groups.

Discussion:

Numerous studies on the methodology of the repair process have raised the question as to whether and to what extent filling materials, which have become very diverse, are compatible. Intermediate agents were proved to have the strongest influence on composite repair bond strengths, whereas pretreatment of the substrate played a minor role.¹⁸

Silanes are adhesion promoters that have two different reactive functional groups that can couple and react with a variety of organic and inorganic components to speed up the union of dissimilar materials.¹⁹ Double-bonded resin composite substrates can react with the organic non-hydrolyzable functional group having a carbon-carbon double bond.^{20,21}

From the results of this study, the null hypothesis was rejected as the experimental factors ("materials" and "storage time") had a significant effect on the repair of SBS. On the other hand, the hypothesis was accepted with the preheating factor as there is no statistically significant difference between repair bond strength of preheated and non-preheated immediately and after storage.

Ormocer is a highly functionalized molecule with a dense network, denser than dimethacrylate polymers. Due to the steric hindrance of the silanized containments of the material, the glass particles, the nanoparticles, and the Ormocer matrix, the double bonds are less reachable during polymerization. As a result, a reduced double bond conversion with a high amount of unreacted double bonds is to be expected.²² Filler size is an important aspect also as 90% percent of

Table 2: Comparison of repair bond strength between Methacrylate & Ormocer groups within preheated (immediate & delayed), non-preheated (immediate & delayed) subgroups

		Methacrylate	Ormocer	P
preheated	immediate	182.96±18.43	155.81±36.73	0.03*
	delayed	168.12±47.58	118.76±34.13	0.008*
non-preheated	immediate	175.58±29.94	181.60±45.88	0.7
	delayed	159.48±32.97	107.65±37.72	0.002*

Data expressed as Mean ±SD, P: Probability, *significance <0.05, Test used: Student's t-test(unpaired)

Table 3: Comparison of Repair Bond Strength between Immediate & groups within Methacrylate (preheated, non-preheated subgroups), Ormocer (preheated, non-preheated subgroups)

		immediate	delayed	P
Methacrylate	preheated	182.96±18.43	168.12±47.58	0.32
	non-preheated	175.58±29.94	159.48±32.97	0.22
Ormocer	preheated	155.81±36.73	118.76±34.13	0.018*
	non-preheated	181.60±45.88	107.65±37.72	<0.001*

Data expressed as Mean±SD, P: Probability, *significance <0.05, Test used: Student's t-test (unpaired)

the fillers in admira Fusion is below 5 µm. This gives less retentive areas after repair treatment; thereby the new composite layer cannot be easily retained.²³

The Nano-hybrid methacrylate-based resin composite used in this study combines 2 different monomers that account for its chemical properties, the large Bis-GMA molecules have higher viscosity and reduced degree of conversion, the smaller and flexible TEGDMA is used as the diluting monomer, increasing the degree of conversion and decrease water sorption.²⁴ It can be also suggested that the bigger filler particles may provide extra retentive areas after surface treatment, thereby the new composite layer can be easily retained. The filler particles of methacrylate are based on silicon dioxide and glass ceramic micro fillers.

Cavalcante et al.²⁵ found that Ormocer like Admira Fusion, containing no dimethacrylates, show higher solubility values, which are independent of the fillers, unlike composites containing dimethacrylates like Grandio.

Intermediate unfilled resins enhance the chemical bond to the matrix and to the exposed fillers, as well as improve micromechanical retention by infiltrating into the micro-irregularities created by the mechanical treatment on the surface Futurabond m +) This could be attributed to the intimate adaptation of the low viscosity resin over the ground resin composite surface and the presence of phosphate groups in FBM+ could contribute to the efficient wetting and bonding to the inorganic fillers in resin composite.3-For ground resin composite surfaces, the use of intermediate adhesive layer could compensate for the loss of unreacted methacrylate groups and render the newly added resin material less viscous and bond to the exposed fillers on the ground surface.

The results were in agreement with Steiner et al.²⁷ study which showed that the type of universal adhesives from the same system of composite could efficiently affect repair bond strength. Elaskary et al.²⁸ showed that Admira Bond is an Ormocer-based adhesive, hence the

chemical adhesion through the monomer chains entanglement between the Ormocer resin of adhesive and the composite. This was not the condition when methacrylate-based resin composite was repaired with Ormocer-based resin adhesive. This might raise the importance of resin matrix compatibility between the adhesive and resin composite in terms of the immediate repair potential of the newly.

The reason for preferring to age the resin sticks by storage in distilled water was that it is an effective and

common method used in previous studies.²⁹ After six months of water storage, it is possible to see from the results that all mechanical parameters decreased as the water absorption increased.³⁰ Water absorption and diffusion are described by two different theories.

The first is known as "free volume theory," and it states that the amount of water that diffuses through microvoids, resin filler interfaces, or morphological defects without connecting to the polar groups of the dimethacrylates composite or Ormocer depends on the free volume fraction of that unbound water.³¹ Water acts as a plasticizer, causing polymer chains to split and decrease the mechanical properties.³² The second theory is "interaction theory," and it states that water binds to polar groups such as ester groups in TEGDMA or hydroxyl groups in Bis-GMA or to the oxygen at the free double bond of the silanized particles of the Ormocer to form -OH bonds. The hydrophilicity of the material affects this process.

Conclusions:

- 1- Immediate and delayed repair bond strength of methacrylate-based resin composite was superior to Ormocer based composite.
- 2- Preheating of methacrylate or Ormocer composite had no effect on immediate and delayed repair bond strength.
- 3- Ormocer based composite groups could not succeed to maintain the repair bond strength after six months of storage, while methacrylate mainly maintains

more than Ormocer.

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