



Micromorphological analysis of HEMA-free versus HEMA-containing adhesive systems bonded to dentin

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ABSTRACT: This research paper aims to investigate the micromorphological pattern of resin/dentin interface of HEMA-free adhesives in comparison with HEMA-containing adhesives. The specimens were examined using scanning electron microscope (SEM) at magnification x1000, half of the specimens were investigated immediately after preparation, while the other half were exposed to 6 months' water storage and 5000 thermal cycles before examination.

KEYWORDS: HEMA-free adhesive; HEMA-containing; micromorphological analysis; composite restorations.

I. INTRODUCTION

Achieving sufficient hybridization inside collagen fibrils and the durability of the resin-dentin interface are both essential for good dentin adhesion, it is important to recognize that, the hybridization mechanism, in which an interdiffusion zone, also known as a "Hybrid layer," is formed, fulfills the occurrence of the micromechanical retention of the restoration.[1]

Due to the relatively high water content of dentin, adhesive compositions should have a specific level of hydrophilicity to adequately wet and penetrate the dentin surface for optimal hybridization. As a result, 2-hydroxyethyl methacrylate (HEMA), which is a hydrophilic monomer, is commonly added to adhesive components, not only as a co-solvent but also as a diffusion promoter for other monomers to form the hybrid layer[2] into the demineralized dentin surface. Besides, HEMA improves the wetting ability of the dentin, because of its high hydrophilicity. [3]

HEMA is considered a possible component that reduces the durability of adhesion, because of water sorption and hydrolytic bond degradation that is associated with HEMA-containing adhesives, [4] this hydrolytic degradation of the adhesive interface has negative clinical effects, including dentin hypersensitivity, marginal discoloration, and the possibility of secondary caries. As a result the durability and stability of the

restorations would reduce.[5] Furthermore, HEMA may be liberated from the adhesive and move through the dentinal tubules to the dental pulp. Thus, it is known with its harmful cytotoxicity and genotoxicity.[6]

For that reason, adhesive systems without HEMA monomer have been introduced to avoid its negative effects.[3] They known as HEMA-free adhesives. However, the lack of HEMA in the adhesive makes it more susceptible to a separation phase between hydrophobic and hydrophilic components.[7, 8] The phase-separation of HEMA-free adhesives would form water droplets by water separation from the adhesive monomer,[9] which frequently expresses water-tree nanoleakage that formed inside the polymerized adhesive layer, because of osmotic infiltration of remaining water on the dentin surface or water transferred from the dentinal tubules.[8, 10]

Although, many studies have been accomplished to assess the laboratory and clinical performance of these adhesive systems, the efficacy of HEMA on the success of composite restorations is still debatable. Numerous studies [7, 11-13] found no statistically significant changes between the clinical and laboratory performances of HEMA-free and HEMA-containing adhesive systems. However, other studies [14-16] reported that these two adhesive systems (HEMA-free and HEMA-containing adhesive systems) performed differently in clinical and laboratory conditions. Thus, this study aims to investigate the micromorphological pattern of these two adhesive systems.

II. MATERIALS AND METHODS:

Twelve freshly extracted human molars were obtained from the oral surgery clinic of Faculty of Dentistry, Mansoura University. The reason of extraction was due to periodontal diseases. Any soft tissue or hard deposits were removed from the molars using hand scaler, rubber cups, pumice water slurry and low speed. The extracted molars were checked using stereomicroscope (ZEISS Stemi



508, Wetzlar, Germany) to detect caries, cracks, and previous fillings.

Only sound, intact and unrestored teeth were chosen for this study. The molars were placed in a 0.5% solution of chloramine-T (Faculty of Pharmacy, Mansoura University, Egypt) for two days to disinfect,[17] then kept in distilled water at 4°C until use. The occlusal enamel was removed using periapical radiographs to reach to the mid-coronal dentin. Poly vinyl chloride (PVC) (PVC 1.4 x 2.5 cm) tubes used as molds where the sectioned teeth were positioned and inserted in cold cured acrylic resin (Acrostone, Egypt). In order to establish a standardized smear layer, the molars' occlusal surfaces were polished for 60 seconds with 600-grit silicon carbide paper.

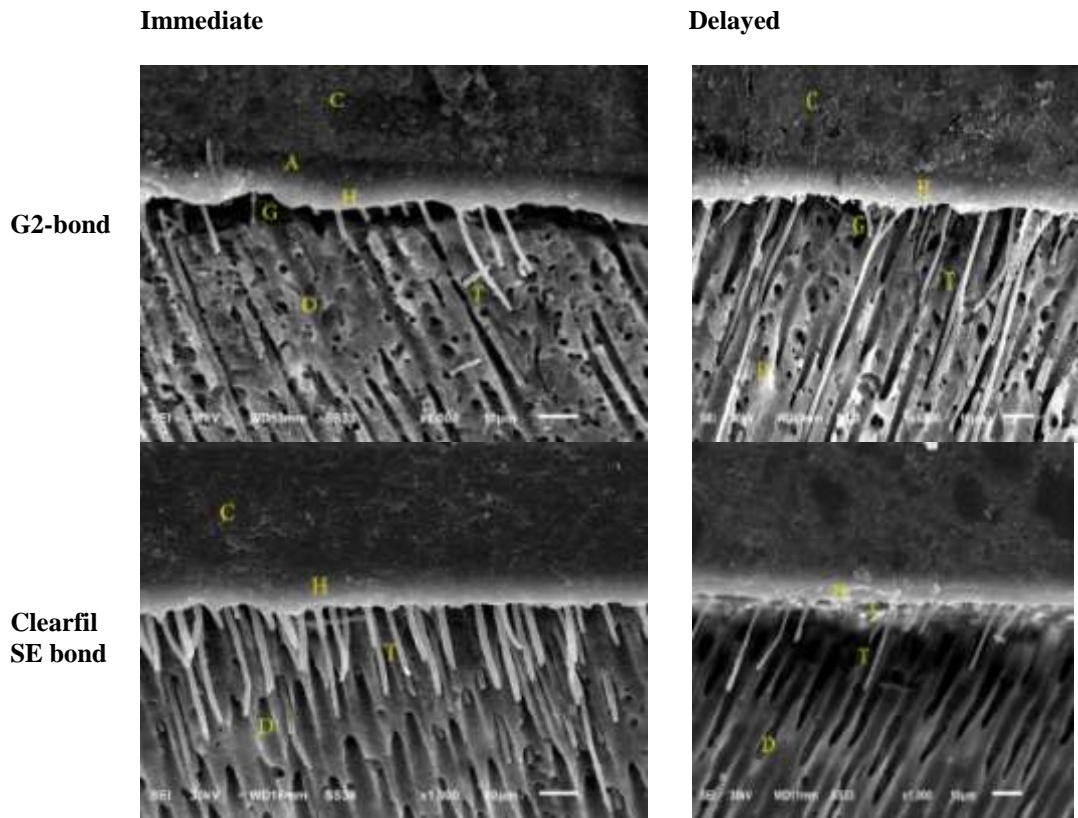
Two dissimilar adhesive systems were applied to dentin surface of the prepared teeth, half of the specimens received HEMA-free adhesive system (G2-bond universal, GC, Tokyo, Japan), and the other half received HEMA-containing adhesive system (Clearfil SE bond, Kuraray Noritake, Tokyo, Japan). Clearfil AP-X (Kuraray Noritake, Tokyo, Japan) resin composite was incrementally placed on the bonded surface in (2-3) ml using a metal band surrounding the specimen and acting as a mold. Each layer was light cured for 20 seconds using a light cure unit (Elipar TM Deep Cure-S LED Curing Light). The intensity of the LED unit was measured by a radiometer with a power of 1300 W/cm and a wavelength range of 350-520 nm. After the composite had fully cured, the metal band was removed, and the teeth were kept at room temperature in distilled water for 24 hours before the test. Half of the specimens were investigated immediately and the other half were assigned to aging protocol which was storage in artificial saliva for 6 months, then thermal cycling for a total number of 5000 cycles.

The teeth were additionally prepared for SEM. Each specimen was divided into two equal halves, using a water-cooled diamond disk at low speed (IsoMetTM 4000, Buehler Ltd., Lake Bluff, LL, USA) along their long axis, perpendicular to the resin-dentin interface. Resin-dentin slabs were obtained from each half and polished with silicon carbide paper of different grits (600, 1000, 1200,

2000 and 4000) using a polishing cloth. The final polish was achieved with diamond pastes of decreasing sizes (6 µm, 4 µm and 1 µm respectively). The specimens were placed in a digital ultrasonic bath (Guilin, Woodpecker, Guangxi, China) to remove the debris. The specimens received a 10-minute ultrasonic cleaning, then kept in saline solution for 10 minutes at room temperature before undergoing an acid-base challenge. This involved exposing them to a 10% orthophosphoric acid solution for 10 seconds and then to a 5% sodium hypochlorite solution for 5 minutes. This technique removed any dentin that was not filled with resin. The specimens were gold sputtered twice (SPI Module - Sputter Carbon / Gold Coater, EDEN instruments, Japan) and examined in secondary electron detection mode using a SEM (JSM- 6510LV, JEOL, Japan) with an accelerating voltage of 30 KV and a working distance of 10-15 mm. The images were taken at magnification x 1000. The methodology followed the published paper by Hamama et al.[18]

III. RESULTS:

A descriptive micromorphological analysis of the interfaces between different adhesives and the dentin surfaces was carried out for each group. SEM images of the G2-immediate group showed tubular penetration of adhesive into the dentin surface, and the formation of discrete resin tags of the adhesive penetrating the dentin surface with the presence of interfacial gaps that represent the smear layer. There were signs of separation at the interface as. The SEM images of Clearfil SE bond immediate group showed long and thick resin tags with funnel shaped pattern penetrating the dentin surface without any evidence separation or interfacial gaps, the presence of a hybrid layer was also obvious. According to the aging group of G2- bond, there were resin tags varied in shape and length penetrating dentin surface, minor interfacial gaps and a thin hybrid layer could be seen. In SEM images of the Clearfil SE aging group there were short and distinct resin tags penetrated the dentin surface with the presence of minor interfacial gaps. (**Figure 1**).



C; composite, A; adhesive, D; dentin, H; hybrid layer, T; resin tags, G; interfacial gaps

Figures 1: SEM micrographs showing the resin-dentin interface of all the tested groups at magnification x1000.

IV. DISCUSSION

Despite the significant advancements in adhesive technology over the past 50 years, many challenges with the longevity of the adhesive interface remain unsolved. It's still difficult to achieve an impervious seal between resin and dentin substrate.[1] Dental adhesion process mainly depends on the production of an appropriate hybrid layer. The creation of hybrid layer mainly depends on the incorporation of monomers within the dentin substrate, regardless of the thickness of the adhesive layer and the depth of the resin tags that penetrate into the dentinal tubules.[19] Therefore, a perfect interdiffusion of the adhesive system inside a collagen scaffold may not be created in a suitable manner.[20]

scanning electron microscope SEM was used to determine the micromorphological topography in the current study because it has high magnification and one of the best tools to determine the adhesive interface between the tooth substrate and dental material, and it is used to show hybrid layer that produced when a monomer is impregnated into the surface of demineralized dentin to create an acid-resistant layer of resin-

reinforced dentin.[21] The immediate group of G2-bond showed little resin-interdiffusion with discrete and few resin tags, signs of separation throughout the interface and interfacial gaps that represent the smear layer can be seen too. Smear layer is a region of tooth preparation debris that is dispersed over the surface during tooth preparation. Some of this debris create smear plugs, which reduce the permeability of the dentin by 86%, by obstructing the orifices of the dentinal tubules[22] and decrease resin penetration. The possible justification might be due to the hydrophobic nature of G2-bond as its HEMA-free adhesive with lower ability for monomer interdiffusion in the hydrophilic dentin. On the other hand, On the other hand, the immediate group of Clearfil SE had more resin interdiffusion with long, thick and funnel shaped resin tags. There was no signs of separation through the interface or interfacial gaps that represent smear layer. This may have attributed to hydrophilic nature of HEMA-containing SE bond.

The aging group of G2-bond, which was exposed to 6-month storage in artificial saliva and 5000 thermalcycles, had more resin interdiffusion than its immediate analogue. A study by Hatirli et



al[23] agreed with this outcome, and showed thermal aging did not affect negatively the bond strength of the adhesive. This might be due to the composition of G2-bond as it contains MDP monomer, thus the aging conditions can hardly degrade it. On the other hand, the aging group of SE bond had lower monomer interdiffusion than its immediate analogue, because sorption and solubility during storage are phenomena that lead to chemical changes, which cause adverse effect on the mechanical properties of polymeric materials, and they also play a role in the hydrothermal degradation of resin composites. Based on the composition and microstructure of the materials, the diffusion of solvents into the polymer network causes a volumetric expansion as a result of the separation of polymeric chains. Aqueous solvent absorption causes swelling, which is accompanied by the loss of non-reacted components, erosion of the filler-matrix interface, and plasticization with a decrease in stiffness, hardness, wear resistance, and flexural strength.[24]

V. CONCLUSION

Additional clinical studies are needed to determine the durability of the restorations bonded with HEMA-free adhesive systems, because the study is limited to laboratory conditions, but within the limitations of this study, it can be concluded, HEMA-free adhesive systems had lower rate of monomer interdiffusion inside dentin, while HEMA-containing adhesive systems had higher rate of monomer interdiffusion.

REFERENCES

- [1]. Hardan L, Bourgi R, Kharouf N, Mancino D, Zarow M, Jakubowicz N, et al. Bond Strength of Universal Adhesives to Dentin: A Systematic Review and Meta-Analysis. *Polym J*.2021;13.
- [2]. Bacelar-Sá R, Sauro S, Abuna G, Vitti R, Nikaido T, Tagami J, et al. Adhesion Evaluation of Dentin Sealing, Micropermeability, and Bond Strength of Current HEMA-free Adhesives to Dentin. *J Adhes Dent*. 2017;19:357-64.
- [3]. da Silva TSP, de Castro RF, Magno MB, Maia LC, Silva ESMHDJ. Do HEMA-free adhesive systems have better clinical performance than HEMA-containing systems in noncarious cervical lesions? A systematic review and meta-analysis. *J Dent*. 2018;74:1-14.
- [4]. Tauscher S, Angermann J, Catel Y, Moszner N. Evaluation of alternative monomers to HEMA for dental applications. *Dental materials* : official publication of the Academy of Dental Materials. 2017;33:857-65.
- [5]. Betancourt DE, Baldion PA, Castellanos JE. Resin-Dentin Bonding Interface: Mechanisms of Degradation and Strategies for Stabilization of the Hybrid Layer. *Int J Biomater*. 2019;2019:5268342.
- [6]. Gallorini M, Cataldi A, di Giacomo V. HEMA-induced cytotoxicity: oxidative stress, genotoxicity and apoptosis. *Int Endod J*. 2014;47:813-8.
- [7]. Tichy A, Hosaka K, Yang Y, Motoyama Y, Sumi Y, Nakajima M, et al. Can a New HEMA-free Two-step Self-etch Adhesive Improve Dentin Bonding Durability and Marginal Adaptation? *J Adhes Dent*. 2021;23:505-12.
- [8]. Mahdan MH, Nakajima M, Foxtom RM, Tagami J. Combined effect of smear layer characteristics and hydrostatic pulpal pressure on dentine bond strength of HEMA-free and HEMA-containing adhesives. *J Dent*. 2013;41:861-71.
- [9]. Torkabadi S, Nakajima M, Ikeda M, Foxtom RM, Tagami J. Bonding durability of HEMA-free and HEMA-containing one-step adhesives to dentine surrounded by bonded enamel. *J Dent*. 2008;36:80-6.
- [10]. Yuan Y, Shimada Y, Ichinose S, Sadr A, Tagami J. Effects of dentin characteristics on interfacial nanoleakage. *J Dent Res*. 2007;86:1001-6.
- [11]. Burrow MF, Tyas MJ. Comparison of two all-in-one adhesives bonded to non-carious cervical lesions--results at 3 years. *Clin Oral Investig*. 2012;16:1089-94.
- [12]. Kubo S, Yokota H, Yokota H, Hayashi Y. Two-year clinical evaluation of one-step self-etch systems in non-carious cervical lesions. *J Dent*. 2009;37:149-55.
- [13]. Moretto SG, Russo EM, Carvalho RC, De Munck J, Van Landuyt K, Peumans M, et al. 3-year clinical effectiveness of one-step adhesives in non-carious cervical lesions. *J Dent*. 2013;41:675-82.
- [14]. van Dijken JW. A 6-year prospective evaluation of a one-step HEMA-free self-etching adhesive in Class II restorations. *Dent Mater*. 2013;29:1116-22.
- [15]. Takahashi M, Nakajima M, Hosaka K, Ikeda M, Foxtom RM, Tagami J. Long-term evaluation of water sorption and ultimate tensile strength of HEMA-



- containing/-free one-step self-etch adhesives. *J Dent.* 2011;39:506-12.
- [16]. Häfer M, Jentsch H, Haak R, Schneider H. A three-year clinical evaluation of a one-step self-etch and a two-step etch-and-rinse adhesive in non-carious cervical lesions. *J Dent.* 2015;43:350-61.
- [17]. Daneshkazemi P, Ghasemi A, Daneshkazemi A, Shafiee F. Evaluation of micro shear bonding strength of two universal dentin bondings to superficial dentin by self etch and etch-and-rinse strategies. *J Clin Exp Dent.* 2018;10:e837-e43.
- [18]. Hamama HH, Yiu CK, Burrow MF. Effect of chemomechanical caries removal on bonding of self-etching adhesives to caries-affected dentin. *J Adhes Dent.* 2014;16:507-16.
- [19]. Iliev G, Hardan L, Kassis C, Bourgi R, Cuevas-Suárez CE, Lukomska-Szymanska M, et al. Shelf Life and Storage Conditions of Universal Adhesives: A Literature Review. *Polymers.* 2021;13.
- [20]. Stape THS, Tjäderhane L, Abuna G, Sinhoreti MAC, Martins LRM, Tezvergil-Mutluay A. Optimization of the etch-and-rinse technique: New perspectives to improve resin-dentin bonding and hybrid layer integrity by reducing residual water using dimethyl sulfoxide pretreatments. *Dent Mater.* 2018;34:967-77.
- [21]. Pereira CN, Daleprane B, Barbosa PF, Moreira AN, de Magalhães CS. Qualitative evaluation of scanning electron microscopy methods in a study of the resin cement/dentine adhesive interface. *Microscopy and microanalysis : the official journal of Microscopy Society of America, Microbeam Analysis Society, Microscopical Society of Canada.* 2014;20:268-75.
- [22]. Saikaew P, Sattabanasuk V, Harnirattisai C, Chowdhury A, Carvalho R, Sano H. Role of the smear layer in adhesive dentistry and the clinical applications to improve bonding performance. *Jpn Dent Sci Rev.* 2022;58:59-66.
- [23]. Hatirli H, Yerliyurt K. Effect of Clinically Relevant Smear Layers and pH of Universal Adhesives on Dentin Bond Strength and Durability. *J Adhes Dent.* 2022;24:87-94.
- [24]. Benalcázar Jalkh EB, Machado CM, Gianinni M, Beltramini I, Piza M, Coelho PG, et al. Effect of Thermocycling on Biaxial Flexural Strength of CAD/CAM, Bulk Fill, and Conventional Resin Composite Materials. *Oper Dent.* 2019;44:E254-e62.