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HEMA-Free Vs HEMA Containing Universal Adhesives: Microtensile Bond Strength, Nanoleakage Patterns and Micromorphological Analysis of Tooth/Restoration Interface; Review literature.

Ali Hadi Qershi^{1*}, Hamdi H. H. Hamama, Salah Hasab Mahmoud¹

¹ Conservative Dentistry Department, Faculty of Dentistry, Mansoura University, Mansoura, Egypt.

*Corresponding author: Ali Hadi Qershi

Email: draliqershi@yahoo.com

Mobile no.: 00201551507460

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Modern adhesives typically include an acidic conditioner, dentin primer, and bonding resin, with a trend toward simplification for universality in dental restorations.¹ New adhesive systems aim to enhance bond strength and durability, with the challenge of selecting the best adhesive system due to the wide variety available.² Key component HEMA has been used to improve wettability since the 1970s, but recently HEMA-free systems have been developed to address certain issues.³

Histology of Dentin

Approximately 50% inorganic particles, 30% organic matter, and 20% water make up dentin. The mineral composition is improved by carbonate and calcium apatite. Collagen type I makes up the majority of its organic component, with trace levels of phosphoproteins, glycoproteins, and proteins containing carboxyglutamate. The transit of the dentinal tubules from the pulp cavity to immediately below the dentin-enamel

junction (DEJ) or dentin-cementum junction (DCJ) is one of the characteristics that set dentin apart.⁴

Between the odontoblastic cells in the pulp chamber and the associated connections, these tubules create tunnels. Where they are will affect their density. It is highest close to the pulp chamber and lowest at the DEJ. A highly mineralized peritubular dentin composed of carbonate apatites encircles dentinal tubules. Perpendicular, unmineralized collagen fibrils and cementum junctions (DCJs) make up intertubular dentin.⁵ It is essential to comprehend the composition of dentin to improve bonding, as it is influenced by its position in the tooth, age, and the presence or absence of disease.⁶

Mechanisms of Adhesion

Adhesives utilize certain functional monomers, such as 10-MDP, 4-MET, and phenyl-P, to create chemical bonds. These monomers have phosphate and carboxylic groups that form ionic bonds with calcium in hydroxyapatite (HAp). The adhesion decalcification concept (AD concept) proposes that when acids come into contact with hydroxyapatite (HAp), they first create ionic connections with the calcium, maintaining an electro-neutral surface. The bond's stability relies on the.⁷

Creation of the calcium salt. Nano-layering, which involves combining 10-MDP monomers and hydroxyapatite in a unique configuration, strengthens the binding between resin and dentin by increasing its resistance to hydrolysis. This, in turn, increases the overall durability of the bond.⁸

The foundation of all adhesive systems now employed is dentin hybridization. This procedure entails the adhesive resin permeating the collagen fibril network that is revealed through acid etching. It replaces the mineral component of the dentinal tissue at the molecular level. The outcome is the creation of a hybrid layer, which is a bonding structure that is distinct from both dentin and resin. The composition of this layer is heterogeneous, consisting of 70% resin and 30% collagen.⁹ Several factors, such as the composition and morphology of dentin, the type of material used, and the technique employed, might influence the subsequent changes in the form and appearance of the hybrid layer.

The systematic review by Carrilho et al.¹⁰ evaluated the performance of 10-MDP-based dental adhesives, emphasizing their chemical interaction with enamel and dentin. The study characterized the adhesive interface and assessed its long-term stability. The 10-MDP monomer exhibits a strong chemical affinity for calcium in hydroxyapatite, forming stable MDP-calcium salts that enhance adhesion. This chemical bond surpasses micromechanical retention alone. Additionally, 10-MDP creates an acid-base resistant zone at the adhesive interface, protecting dental tissues from acidic challenges and ensuring bond stability under fluctuating pH conditions in the oral environment.

Classification of Adhesive Systems and Their Application Techniques

1) Etch-and-Rinse adhesives

The etch-and-rinse (ER) mode of application is known to be done only as a multi-step technique, unlike the other approaches. The ER mode is characterized by a mandatory separate etching step by an etchant that

possesses the highest acidity among all the other etching techniques, followed by a rinsing step.¹¹ leading to the complete dissolution of the smear layer, utilizing acid etchant materials enhances the quality of the resin tags formation by enabling them to infiltrate the dentinal tubules with no obstacles effectively.¹²

Despite being the gold standard due to their highest instant bond strength compared to all of the other systems,¹³ ER approach promotes hydrolysis and degradation of the resin-dentin interface over time attributed to its technique and the Existence of water residue in the dentin.¹⁴

2) Self-Etch Adhesives

Self-etch (SE) mode of application is known for eliminating separate etching steps and being satisfied with its own acidic functional monomer abilities to facilitate the infiltration of the monomers through the dentinal tubules. Due to the low acidity of the functional monomer compared with the etchant used within the ER approach, the acidic monomer tends to only modify the smear layer rather than complete dissolution leaving areas of intact smear layer with areas of porous smear layer allowing for some amount of resin tags penetration as well as preserving the highest available amount of the hydroxyl apatite for the future chemical adhesion.¹⁵ Given to the elimination of rinsing step, the dissolved smear layer is mixed with the hybrid layer, and the need to control the degree of moist is no longer required, therefore considering the SE approach as less technique sensitive than ER.¹⁶

3) Universal (Multi-Mode) Adhesives

Researchers and practitioners have recently focused their attention on a new type of dental adhesives known as universal adhesives (UA). The

notion of a "universal adhesive" is ambiguous and somewhat disputed. They claim to function for both direct and indirect restorations and are compatible with self-cure, light-cure, and dual-cure resin-based cement.¹⁷

Universal adhesive necessitates the utilization of adaptable monomers capable of interacting with diverse materials, amalgamating with resin-based restoratives and cement, and possessing a hydrophilic characteristic to effectively wet dentin. They must be thin enough to avoid interference with indirect restorations and acidic enough for self-etching without breaking down polymerization initiators. However, they should still have enough acidity to allow for self-etching without compromising the integrity of polymerization initiators.¹⁸

Universal adhesives should not be confused with 1-step self-etching single-bottle or "all-in-one" systems. These adhesives possess unique chemical compositions, which consist of carboxylate or phosphate monomers. One of the most commonly utilized monomers is 10-methacryloyloxydecyl dihydrogen phosphate (10-MDP).¹⁹ Universal adhesives can consist of many functional monomers, such as glycerol phosphate dimethacrylate (GPDM), 4-methacryloxyethyl trimellitic acid (4-MET), and 4-methacryloxyethyl trimellitate anhydride.²⁰

A systematic review and meta-analysis conducted by Dreweck et al.²¹ evaluated the efficacy of gold standard adhesives compared to various universal adhesives. The findings indicated that these universal adhesives exhibit a similar level of performance to the gold standard adhesive.

Challenges in Dentin Adhesion

The adhesion to dentin continues to pose a hurdle to the clinical outcomes anticipated of these materials, despite an increasing number of

bonded restorations with resin-based materials and advancements in the composition and processes of dental adhesives.²² The particular properties of this complex tissue, including hydrated structural heterogeneity, composition fluctuation, and specialized structure-property linkages, are tightly linked to the dentin substrate and its bonding performance and degradation. Furthermore, several dentin substrates are generated by physiological, aging, and pathological processes that modify dentin. This variability increases the likelihood of failure or degradation of the resin-dentin bond.²³

The primary reason for failure is believed to be the degradation of the hybrid layer at the dentin-adhesive interface. The outcome is the result of the interaction between several mechanical and chemical components, including the hydrolysis and enzymatic degradation of the exposed collagen and adhesive resin. Collagen fibrils can degrade either just after the bonding agent is applied or after the adhesive component has polymerized.²⁴

Smear Layer

The smear layer is a zone of dental preparation material that appears to spread over the surface during tooth preparation. A portion of this waste gathers in the openings of the dentinal tubules, creating obstructions that decrease the permeability of dentin by around 86%. Nevertheless, the microporous structure of the smear layer facilitates the permeation of fluid from the dentinal tubules. The thickness varies between 0.5 and 2 μm . The smear layer consists mainly of hydroxyapatite and collagen, which have undergone denaturation due to friction and heat during tooth preparation.²⁵

However, it has been shown that ER²⁶ adhesive causes a more pronounced and lasting degradation of dentin collagen compared to self-etching adhesive. To prevent the collapse of collagen fibrils, it is important to keep the acid-demineralized dentin moist. However, it is crucial not to make the dentin too wet, since this will hinder the resin impregnation of the collagen fibrils.⁹

Self-etching adhesives have been claimed to be user-friendly due to their simple application and lower technique sensitivity compared to ER adhesives.²⁷ Without phosphoric acid etching, the smear layer is not entirely removed and the remaining portion is incorporated into the hybrid layer. The self-etching adhesives are categorized into four degrees based on their acidity: strong (pH ≤ 1), intermediately strong (pH 1-2), mild (pH ~ 2), and ultra-mild (pH > 2.5). The etching capacity of the adhesives is directly proportional to their pH.²⁸

Degradation at the Resin-Dentin Interface

The degradation and the dissolution of collagen fibrils, along with the activation of naturally occurring proteases called extracellular matrix metalloproteinases (MMPs) and dentin-resident cysteine cathepsins (CTs), are the specific explanations for the degradation at the bonding contact. MMPs and CTs function as collagen-degrading enzymes, specifically targeting the organic matrix of demineralized dentin and resulting in the degradation of the hybrid layer.²⁹

This enzymatic breakdown is associated with the hydrolysis and leaching of the adhesive from the spaces between the fibrils. Hydrolytic degradation refers to the breaking of covalent bonds between polymers when exposed to water. Additionally, it is believed to be the primary factor contributing

to the deterioration of the hybrid layer.³⁰ Due to dentin's hydrophilic nature, the adhesive needs to form a hybrid layer by enabling a hydrophilic resin monomer to penetrate the moist, demineralized dentin.³¹

Strategies to Reduce Hybrid Layer (HL) Degradation

Enzymes Inhibitors

Chlorhexidine (CHX)

Chlorhexidine is the most extensively researched enzymatic inhibitor and antibacterial agent. It is particularly notable for its exceptional capacity to hinder MMP activity in dentin and resin. Due to its ability to chelate at low concentrations (0.2%), it showed excellent suppression of protease enzymes. The binding of CHX to both mineralized and unmineralized dentin is likely to be responsible for its cationic activity. However, there has been limited research conducted on the compound's mode of action. CHX, being composed of chlorine, forms a bond with the zinc present in the catalytic domain of MMP enzymes, hence impeding its hydrolytic activity. MMPs are enzymes.^{32 33}

Quaternary ammonium methacrylates compounds (QAMS)

QAMS has the capacity to inhibit dentinal matrix metalloproteinases (MMPs), hence maintaining the stability of the adhesive bond over a period of time. Quaternary ammonium compounds exhibit strong antibacterial activity when they are positively charged at pH levels that are conducive to good health. These compounds are positively charged and can dissolve in water, similar to CHX. However, they may not seep out of surfaces that they are attached to. QAMs effectively prevent the degradation of demineralized dentin collagen by MMP enzymes.²⁴

Others Enzymatic Inhibitors

Ethylene diamine tetra acetic acid (EDTA)

Ethylene diamine tetra acetic acid, an ethylene-diamine tetra acetic acid, has been used in endodontics for many years due to its ability to form complexes with metal ions. EDTA removes calcium ions (Ca^{2+}) from the collagen matrices by binding to zinc ions (Zn^{2+}) found in the catalytic site of MMPs. Nevertheless, the primary drawbacks include a protracted application duration and the potential for reversibility due to water solubility. Studies have shown that a concentration of 17% weight of EDTA effectively prevents the activity of MMPs induced by self-etching adhesives. Nevertheless, the anti-MMP effects of EDTA are widely acknowledged. Enzymes that are recognized and identified.³⁴

Microtensile Bond Strength Test

Microtensile Bond Strength testing offers several advantages over standard bond strength testing. One advantage is the reduced occurrence of faults due to the small specimen size, often 1 to 2mm or less. Additionally, μTBS testing allows for several measurements to be obtained from a single tooth. The μTBS test is the most efficient method for assessing adhesive retention. To evaluate the lifespan of adhesive restorations, it is necessary to subject the specimens to a durability test. This test was first introduced by Sano et al.³⁵ in 1994.

Noppawong et al.³⁶ Evaluated The microtensile binding strength of three universal adhesives to dentin contaminated with a hemostatic material. And concluded The microtensile bond strength (μTBS) of the

universal adhesives in SE mode was significantly reduced when the dentin was contaminated with a hemostatic substance. However, the detrimental effect was not noticed when the universal adhesives were used in emergency reaction mode.

Nanoleakeage Pattern Test

The structural stability of the hybrid layer may be compromised when water and organic solvents are present in the adhesive resin higher levels of acetone in adhesives used for individual bottles result in reduced strength of microtensile bonds and the formation of gaps and cracks on the surfaces being bonded.³⁷ Ammoniacal silver nitrate and scanning electron microscopy (SEM) were used as observable tracers to investigate the leakage of bonded surfaces. Additionally, water diffusion was observed. The tracer stains the water-filled infiltration routes along the bonded interface, commonly known as "water-trees". These water channels are distinct and extend from the hybrid layer into the adhesive layer.³⁸

Adhesives with high acidity levels experience hydrolytic instability. Nanoleakage occurs due to the presence of nano-porous regions inside the hybrid layer. Water can gradually penetrate via the nanoleakage of the bonded layer, even if it is initially minimal. The decrease in bond strength may be attributed to the plasticization of the resin matrix and the leaching of unreacted monomers caused by the passage of water through the bonded interface.³⁹

Araújo-Neto et al .⁴⁰ Investigated the effects of replacing HEMA with glycerol-dimethacrylate on specific physicochemical properties and the effectiveness of basic adhesive models. And Scientists have found that glycerol-dimethacrylate, a hydrophilic dimethacrylate monomer, can be utilized instead of HEMA in simpler adhesives to enhance dentin adhesion,

mechanical properties, polymerization, and solubility while reducing water absorption.

Micromorphological analysis of the adhesive interface

This provides important information on the adhesive performance of the material, as well as a qualitative evaluation of the adhesive interface through investigation using a scanning electron microscope. Due to the superior clarity and significant magnification of the scanning electron microscope images of HL, this testing method has become widely used for studying the process of adhesive bond formation. For SEM examination, specimens must undergo a process of gold coating. Micrographs were captured with magnifications ranging from 10x to 50000x. While this test offers numerous benefits, it also presents significant disadvantages. Specimen dryness and typical occurrence of de-bonding of the adhesive/dentin interface due to specimen dryness and collagen matrix shrinkage.^{41, 42}

Shin et al.⁴³ The study investigated the impact of several surface treatments on mineral trioxide aggregate (MTA) and their microstructural changes on the bonding strength to composite resin utilizing four adhesive techniques. The acidic treatment had an impact on the micromorphology of the MTA surface and its binding strength to the composite. Utilizing a one-step SE adhesive approach in placing a composite resin restoration over MTA cement can result in a robust bond to WMTA, as observed in this study.

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