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Research progress in dental bonding technology

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Abstract: Dentin bonding is a core technique in oral clinical practice. It seals the dentin tubules through mechanical interlocking and chemical bonding, isolates external stimuli, enhances retention, reduces microleakage, decreases postoperative sensitivity, and the risk of secondary caries, while dispersing stress to protect the pulp. It serves as the foundation for the success of modern dental restoration. This article provides a review of the mechanisms and influencing factors of dentin bonding, as well as methods to improve bonding performance. It discusses the latest research progress in dentin bonding in recent years, aiming to provide references for the development of more durable and stable dentin bonding materials and techniques.

Keywords: Dentin; Adhesive; Bonding Mechanism; Bonding Durability

Preface

Dentin bonding, as a core technology in the field of oral restoration, directly determines the functionality and long-term stability of restorations. This article will provide a comprehensive review of the influencing factors of bonding, systematically analyze existing technical bottlenecks, and explore the potential and limitations of improved strategies such as ethanol wet bonding, enzyme inhibitors, biomimetic remineralization, and fibrous external demineralization. By integrating research results from multiple disciplines, we aim to promote the development of dentin bonding technology towards greater stability and durability.

1 Factors affecting dentin bonding

1.1 Factors affecting the durability of dentin bonding

Research indicates that the average service life of composite resin restorations is 6 to 7.2 years [1], and secondary caries caused by interfacial degradation is one of the main reasons for bonding failure [2]. In fact, the degradation of two important components in the resin-dentin bonding interface - enzymatic hydrolysis of collagen fibers and hydrolysis of resin monomers - is the core of interfacial failure. The activation of endogenous enzymes, the movement of interfacial water, and the metabolic products of bacteria in the oral microenvironment play important roles in interfacial degradation. These factors work together to gradually destabilize the bonding interface.

1.1.1 Activation of endogenous enzymes

Activation of endogenous enzymes in dentin is one of the primary causes of collagen fiber degradation. The bonding process activates MMPs and CTSs in dentin [3]. The activation of MMPs and CTSs typically occurs in an acidic environment, so the acidic properties of adhesives and the physiological environment of dentin can significantly affect the activity of these enzymes. Therefore, controlling the activity of MMPs and CTSs, or using corresponding inhibitors, may help improve the long-term stability and durability of dentin bonding.

1.1.2 Influence of water molecules on the bonding interface

The inherent characteristics of dentin, the high-humidity oral environment, and the water absorption of hydrophilic monomers in adhesives all contribute to the inevitable presence of water molecules at the dentin bonding interface.

(1) Collagen degradation

During the bonding process, the adhesive cannot fully encapsulate the demineralized collagen fibers. The unencapsulated collagen fibers are prone to degradation, and water is one of the main causes of collagen degradation [4].

(2) Degradation of adhesive resin

Hydrolysis is considered to be the primary factor leading to the degradation of resin materials in the mixed layer [5]. Water molecules not only dilute the concentration of resin monomers, affecting the degree of polymerization, but also directly degrade the hydrolysis-sensitive

groups in the resin monomers, resulting in resin swelling and plasticization, which in turn exposes collagen fibers and accelerates their degradation [5]. The ester bonds in methacrylate adhesives are prone to hydrolysis [6], releasing residual monomers and hydrolysis products.

In addition, prostheses that are exposed to oral saliva for a long time may increase the surface area of exposed adhesive, further accelerating this process.

1.1.3 Interfacial stress effect

When restorations are exposed to the oral environment with repeated changes in occlusal force and temperature for a long time, the dentin bonding interface bears various stress effects, including internal stress generated by the polymerization shrinkage of light-cured resin, internal stress caused by the mismatch in thermal expansion coefficients between composite resin and dentin, and external stress generated by daily chewing. Long-term stress effects can lead to fatigue of the bonding interface, gradually causing gaps and forming nanoleakage, thereby damaging the marginal fitness [7].

1.1.4 Lack of chemical bonding in adhesion

Currently, the widely used dental adhesive monomers (such as Bis-GMA, UDMA, TEGDMA, and HEMA) lack stable chemical bonding with dentin, mainly relying on mechanical interlocking. Therefore, developing monomers that can chemically bond with both hydroxyapatite (HA) and collagen is an important direction for future research.

1.1.5 Bacterial infection

The dentin bonding interface undergoes long-term hydrolysis, enzymatic degradation, and various stress effects, leading to the gradual aging and degradation of the hybrid layer. This provides a pathway for the entry of oral saliva and exogenous cariogenic bacteria. Bacteria produce acidic substances that dissolve hydroxyapatite (HA) and release ions to accelerate interface degradation [8]; in addition, enzymes released by saliva, gingival crevicular fluid, and oral microorganisms can also degrade resin monomers, and the degradation products promote bacterial colonization at the resin-dentin bonding interface [9]. Bacterial colonization leads to the accumulation of a large amount of biofilm, in which bacteria can not only form toxic effects at the interface but also further cause secondary caries, resulting in microleakage at the bonding interface [10]. Furthermore, the acidic environment produced by bacteria can activate endogenous enzymes (such as matrix metalloproteinases (MMPs) and cathelicidins (CTs)) in dentin, further degrading collagen fibers.

1.2 Methods to address the durability of dentin bonding

1.2.1 Ethanol wet bonding

Wet bonding technology prevents collagen fibers from being too close by keeping the dentin moist, which facilitates the penetration of adhesive. However, its technical sensitivity is high. Ethanol wet bonding technology promotes the full penetration of adhesive to form a hydrophobic mixed layer through dehydration using gradient ethanol solutions [11], but the complicated and time-consuming operation limits its clinical promotion.

1.2.2 Improve the conversion rate of adhesives or develop hydrophobic adhesives

Addressing the issue of hydrophilic bonding monomers being prone to hydrolysis, research has proposed the development of non-ester bonding monomers (such as amide monomers) to enhance hydrolysis resistance [12]. These strategies contribute to improving the stability and durability of the resin-dentin bonding interface.

1.2.3 Application of MMPs inhibitors

Considering the role of MMPs in collagen degradation, exogenous MMPs inhibitors have begun to attract the attention of numerous scholars. Common MMPs inhibitors in research include chlorhexidine, tetracyclines, and quaternary ammonium compounds. These inhibitors effectively inhibit the activity of MMPs by altering the structure of MMPs protein molecules and interfering with the activation process of MMPs on demineralized collagen matrices. To a certain extent, this reduces collagen fiber degradation and enhances the stability of dentin bonding.

1.2.4 Application of crosslinking agent

Exogenous chemical, physical, or biological crosslinking agents can be used to modify the molecular structure of collagen, enhance its

mechanical stability, and inactivate the active sites of collagenase. Crosslinked collagen-based materials have been widely used in the biomedical field[13].Common crosslinking agents include glutaraldehyde, carbodiimide, and polyphenolic crosslinking agents. These crosslinking agents can improve the durability of dentin bonding and reduce the degradation rate of collagen fibers to some extent by increasing the degree of crosslinking of collagen fibers. However, these methods cannot fundamentally solve the problem of collagen fiber exposure at the bottom of the mixed layer.

1.2.5 Remineralization of the bonding interface

Utilizing biomimetic remineralization technology to fill the collagen-exposed area at the bottom of the mixed layer is considered an ideal method to enhance interfacial stability. Some studies have successfully replaced water molecules in areas with poor adhesive penetration with HA crystals by inducing the ordered deposition of HA within demineralized collagen fibers [14]. This process not only keeps MMPs in an inactive state but also restores the natural mineralization state of collagen fibers under the encapsulation of inorganic mineral crystals, effectively protecting the collagen fibers and restoring their mechanical properties.

Currently, non-collagen analogues are commonly used to induce mineralization, such as carboxyl-modified polyamide amines, polyacrylates, and artificial peptides. These substances promote the mineralization of collagen fibers by mimicking the natural mineralization process.

However, despite the progress made in these remineralization studies in vitro simulations, the process of inducing mineralization is slow and the conditions are demanding. More importantly, collagen fibers may be degraded before they are mineralized, leading to unsatisfactory results. In addition, the oral environment is a complex multifactorial system, and there is still a significant gap between remineralization methods under laboratory conditions and clinical applications.

1.2.6 Fiber external demineralization

“Fiber demineralization” aims to remove only the minerals between collagen fibers while retaining the minerals within the collagen fibers. This provides penetration space for the bonding monomer, while maintaining the integrity of the collagen fibers, preventing their collapse, avoiding MMPs activation, and reducing the risk of collagen fibers being enzymatically degraded [15]. In this way, the adhesive can penetrate fully on the dry dentin bonding surface, forming a stable and durable bonding interface, significantly reducing technical sensitivity and providing new ideas for dentin bonding.

Studies have found that during the demineralization process of dentin, the minerals outside the collagen fibers demineralize first, while the minerals inside the collagen fibers demineralize more slowly. Additionally, using cross-linked dextran exclusion chromatography, the study confirmed that the layered arrangement of type I collagen fibers produces a “molecular sieve” effect [16]. Based on this principle, theoretically, by controlling the demineralization process, high molecular weight calcium ion chelators (COO-) can be used to act on the surface of dentin, demineralizing only the HA outside the fibers, thus achieving selective demineralization outside the collagen fibers.

In recent years, research on fiber external demineralization has been continuously deepening. For example, researchers have successfully achieved fiber external demineralization by using high molecular weight calcium ion chelators such as polyacrylate sodium salt, carboxymethyl chitosan, EDTA-chitosan, and EDTA-ethylene glycol chitosan as demineralizers.

2 Discussion

The long-term stability of dental bonding is a core challenge in the field of oral restoration, and its fundamental issue lies in the dynamic degradation process of the bonding interface. This complex process involves a vicious cycle of various factors, such as the continuous activation of endogenous enzymes, hydrolysis of hydrophilic resin monomers, erosion by oral microorganisms, and accumulation of interfacial stress.

Current research progress indicates that addressing this issue requires a comprehensive multi-pronged strategy: 1) developing more efficient MMPs inhibitors; 2) developing multifunctional adhesives with bonding strength, biological activity, antibacterial properties, and hydrolysis resistance; 3) optimizing the clinical application feasibility of remineralization technology; 4) developing novel adhesive systems that combine functional monomers, crosslinking agents, and remineralization technology.

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