The Effect of MTYA•G•H Treatment on the Adhesion between Pulpal Side Dentin and Composite Resin

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In this study we investigate the adhesiveness of composite resin on pulpal side dentin treated with the primer MTYA•G•H as compared with that for labial side dentin. Fresh bovine dentin was used; the labial and pulpal surfaces were prepared as adherends. The etching agents were 40% phosphoric acid, 10% citric acid, or 0.5 M EDTA (pH 7.4) solutions. Tensile bond strengths were measured after immersion for one day in 37°C water. There was no significant difference between the mean bond strength for labial side dentin and that for pulpal side dentin for each etchant. The highest mean bond strength, about 12MPa, was obtained when pulpal side dentin was etched with EDTA. It is concluded that MTYA•G•H treatment of the deep portions of the dentin is effective in obtaining good adhesion.

Key words: Adhesion, MTYA•G•H, Pulpal dentin

INTRODUCTION

With the objective of improving the adhesion between dentin and composite resins, various studies have been carried out aimed at the development of adhesive monomers or improvements in etching agents. In recent years, special attention has been focused on the attempt to enhance the adhesion of resins to dentin by treating the dentin with a dentin primer after etching¹⁾.

It has been reported that GLUMA (5% glutaraldehyde and 35% HEMA aqueous solution)^{2,3)}, 35% HEMA aqueous solution⁴⁾, or NMSA (N-methacryloyl aminosalicylic acid) in acetone, or certain organic solvents^{5,6)} are effective in improving the adhesion between composite resin and dentin. Chigira *et al.*⁷⁾ reported that the application of GM (glyceryl methacrylate) aqueous solution completely prevented the formation of a contraction gap in a cylindrical dentin cavity.

Endo⁸⁾ and Hayakawa *et al.*⁹⁾ prepared a primer composed of MTYA (methacryloyl tyrosine amide), HEMA, and glutaraldehyde, which they abbreviated as MTYA•G•H, and found that the bond strength of composite resin or MMA/TBB-O resin to dentin was markedly increased when the dentin was treated with MTYA•G•H after etching. Improved durability of bond strengths after MTYA•G•H treatment, as determined by thermocycle tests^{10,11)}, as well as clinical application of this primer¹²⁾, have also been reported.

On the other hand, in discussing the strength of bonding of resin to dentin, the type of the tooth used, preserving conditions and adhered sites must be considered¹³⁾. The structure of the collagen constituting the dentin, the size of apatite crystals and nature of the proteins other than collagen vary depending on the above factors, having a great effect on bond strength.

With regard to the effect of the adhered sites of the dentin, bond strengths in the deeper part of the dentin near the dental pulp have been demonstrated to be lower than those in the superficial layers of the dentin¹⁴⁻¹⁹. This may result either from differences in chemical composition or from regional differences in wetness.

The objectives of this study were to investigate the effect of MTYA•G•H treatment on deep dentin adhesion as compared with superficial dentin adhesion. For the superficial dentin on the labial side was studied, while deep dentin on the pulpal side was used. The former has been commonly used in tensile bond tests, and the latter is considered to be distinctly different from the former in characteristic features such as the degree of calcification and the structure of the dentin, including dentinal tubules. In addition, many substances adhere to it with difficulty.

MATERIALS AND METHODS

Preparation of MTYA • G • H

Using MTYA synthesized by the method described in a previous report,²⁰ HEMA,* and glutaraldehyde,** the primer MTYA•G•H was prepared with the composition shown in Table 1. Solutions A and B are mixed well just before use.

Preparation of test samples

Anterior teeth were extracted from the mandibles of bovines just after sacrifice. The teeth were frozen immediately after removal of the coronal pulp, and thawed just before use. Using a grindstone and water spray, the enamel was removed in order to expose the labial and pulpal dentin surfaces as shown in Fig. 1. These dentin surfaces were ground with a # 800 grinder, and then with a # 1000 grinder.

Procedure for adhesion and the test methods for tensile bonds

Figure 1 shows the procedure for adhesion. The etchants used were 40% phosphoric acid, 10% citric acid, or 0.5 M aqueous EDTA solution adjusted to pH 7.4.

After grinding, the labial and pulpal dentin surfaces were etched with the above mentioned agents for 60 s, and thereafter washed and dried. MTYA•G•H was applied to each dentin surface for 60 s. After drying, a silicone rubber ring, 3.2 mm in inside diameter and 2.0 mm deep, was temporarily attached to each surface. The bonding agent* was applied to the inside of the ring, and the ring was filled with composite resin**. The ring was removed after the resin was cured. Each sample was stored in water at 37°C for one day, and then examined for tensile bond strength. A brass attachment was fixed to the upper part of

Table 1 Composition of MTYA • G • H

A : 6% MTYA-70% HEMA aqueous solution
B : 2% glutaraldehyde aqueous solution

* E. Merck, Darmstadt, Germany

** Wako Pure Chemical Industry Ltd., Osaka, Japan

* Clearfil New Bond, Kuraray Co., Osaka, Japan

** Clearfil Core, Kuraray Co., Osaka, Japan

Fig. 1 Procedures for preparing test samples.

the resin, and the bond strength was measured at a cross head speed of 2 mm/min, using an INSTRON universal testing apparatus***. Seven to twelve samples were tested for each given condition.

*** TOM-10000X Shinko Co., Tokyo, Japan

RESULTS

Bond strength on the dentin surface

Figure 2 shows the bond strength of composite resin when adhering to the labial and pulpal dentin surfaces.

The mean value for labial side dentin was about 10 MPa after 40% phosphoric acid etching, about 9 MPa after 10% citric acid etching, and about 7 MPa after EDTA etching.

The effects of the etchants on pulpal side dentin was distinctly different from those observed for labial dentin surfaces. In other words, the bond strength was only about 8 MPa on pulpal dentin surfaces after 40% phosphoric acid etching. In addition, the highest mean bond strength, about 12 MPa, was obtained after etching with EDTA, which is considered to be the mildest among the three etching agents. With citric acid etching, the value was the lowest, about 5 MPa.

The difference in the mean bond strength was analyzed by Scheff's test at 95% confidence limits for each etchant. There was no significant difference between mean bond strength for labial side dentin and for pulpal side dentin.

SEM observation of etched dentin surface

Figure 3 shows SEM pictures of labial dentin surfaces after etching. The surfaces are free of smear and dentinal tubules were opened infundibularly after 40% phosphoric acid etching. After etching with 0.5 M EDTA solution, the smeared layer was removed, but the tubules were obstructed.

SEM pictures of pulpal side dentin after etching are shown in Fig. 4. There was no infundibular dilation of the dentinal tubules after etching with 40% phosphoric acid, unlike the changes in the labial dentin surface, and phosphoric and citric acids caused marked erosion of the dentin. Following etching with 0.5 M EDTA, the smeared layer was obviously removed and dentinal tubules were exposed.

Fig. 2 Tensile bond strength of composite resin on labial side and pulpal side etched dentin.

Fig. 3 SEM of labial side etched dentin.

Fig. 4 SEM of pulpal side etched dentin.

DISCUSSION

The types of dentin and adhered sites are important factors in the adhesion of resin to dentin. Particularly when the nature of the site varies, it has been reported that dentin bond strengths are lower on the pulpal side than on the labial.¹⁴⁻¹⁹⁾ This has been explained by the lower degree of calcification on the pulpal side or by regional differences in wetness. Suzuki *et al.*¹⁸⁾ reported that there is a correlation between the area of the intertubular dentin and the bond strength, and that the bond strength is less at sites proximal to the pulp because of the reduced area of the intertubular dentin. Tagami *et al.*¹⁹⁾²¹⁾ reported that wetness affected

adhesion to deep dentin.

In the present study, labial and pulpal dentin surfaces, which differ from each other in dentinal structure, were compared with regard to adhesiveness after treatment with MTYA•G•H. In bovine teeth, pulpal dentin is less calcified than labial dentin,²²⁾ and the difference between surface conditions as shown in Fig. 3 and 4 results from differences in the degree of calcification.

The highest mean value was obtained when pulpal dentin was etched with EDTA. For the less calcified pulpal side dentin, EDTA etching is adequate to produce intertubular and peritubular dentin which is more likely to allow permeation of MTYA•G•H, resulting in a higher bond strength. For labial side dentin, EDTA pretreatment is too mild to increase bond strengths, as reported previously¹⁰⁾. It has also been reported²³⁾ that acid etching denatured the collagen, affecting the low bond strength. For pulpal side dentin, the degree of collagen denaturation is thought to be higher because of the lower degree of calcification, as mentioned above. However, in this study, MTYA•G•H treatment raised the bond strength not only for labial side dentin but also for pulpal side dentin, when denatured by phosphoric acid or citric acid etching.

The decalcified layer in pulpal side dentin is collagen rich, and MTYA • G • H reacts more readily with that layer, according to the mechanism presented previously.¹⁰⁾¹¹⁾ However, mean bond strengths for pulpal side dentin etched with citric acid are significantly lower than those obtained using EDTA. It is presumed that the difference in the degree of decalcification caused by acid etching is not the only factor affecting adhesiveness, but that other factors are involved such as the characteristic features of collagen after etching, and the reactivity of reaction sites of collagen with glutaraldehyde.

If the formation of resin tags is the main factor contributing to dentin adhesion, the bond strengths observed for labial side dentin after phosphoric acid etching should be the highest, because of the wide dilation of the dentinal tubules. However, there are no differences between bond strengths for labial side dentin after phosphoric acid etching and those to labial after EDTA etching despite great differences in surface conditions, as shown in Fig. 3 and 4. The highest mean value was obtained when pulpal side dentin was etched with EDTA. These results suggest that the main factor in dentin adhesion is not the formation of resin tags but is the interpenetration of monomers into the intertubular and peritubular dentin. MTYA•G•H treatment makes this kind of penetration easier, and yields high bond strengths not only for labial side dentin but also for pulpal side dentin.

CONCLUSION

The effects of MTYA•G•H treatment on the adhesion between composite resin and labial or pulpal dentin surfaces were investigated. It was difficult to obtain good adhesiveness with pulpal side dentin in the absence of MTYA•G•H treatment. MTYA•G•H treatment was apparently effective in increasing the adhesion of composite resin to pulpal side dentin, suggesting that it may also be useful in increasing its adhesion to deeper dentin, such as root canal dentin.

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本号掲載論文の和文抄録

デュアルキュアー型 コンポジットレジンセメントの反応率

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デュアルキュアー型コンポジットレジンセメントの硬化反応率をフーリエ変換赤外吸収分光法で測定した。デュアルキュアー型レジンセメントの反応率は、レドックス反応による常温硬化では 59.3~75.0 %, 光硬化では 66.6~81.4 % を示した。光硬化時の反応率は、セメントと照射器照射口の間に介在するコンポジットレジンインレー体の厚さが増加するにつれて減少した。また、光硬

化時の低反応率は、経時的に十分には向上しなかった。したがって、デュアルキュアー型のレジンセメントを光硬化させる場合、充填用光重合型コンポジットレジンの場合と同様十分光照射しなければならない。反応率を向上させ、均一化するためには、低反応率になったと思われる部位に照射口を近づけて再照射する方法が非常に有効であった。

歯髄側象牙質とコンポジットレジンセメントとの接着に及ぼす MTYA・G・H 処理の効果

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本研究では、歯髄側象牙質を MTYA・G・H で処理したときの接着強さを、唇側象牙質と比較しながら調べた。

新鮮牛抜去歯の唇側、あるいは歯髄側象牙質表面を被着体として用い、40 % リン酸, 10 % クエン酸, または 0.5 M の EDTA 水溶液でエッチングし、37°C 水中に 1 日浸漬後の引張接着強さを測定した。

各エッチング剤間で、唇側象牙質と歯髄側象牙質とに

対する接着強さを比較したところ、有意な差はみられなかった。また、歯髄側象牙質を EDTA エッチングした場合に、平均値で約 12 MPa と最も高い値が得られた。

以上の結果より、深部象牙質に対して、MTYA・G・H 処理を施すと良好な接着強さが得られることが判明した。

歯科用接着性モノマー (4-methacryloyloxyethoxycarbonylphthalic Anhydride, 4-META) の溶血性と NMR, DSC による 4-META/りん脂質リポソームの相互作用

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