Durability of Adhesion between 4-META/MMA-TBB Resin and Cementum

Yuko TANAKA, Tsutomu SUGAYA and Masamitsu KAWANAMI

Periodontology & Endodontology, Department of Oral Health Science, Hokkaido University Graduate School of Dental Medicine, Kita 13 Nishi 7, Kita-ku, Sapporo, 060-8586, Japan
Corresponding author, E-mail:yuuko@den.hokudai.ac.jp

4-META/MMA-TBB resin has shown good biocompatibility and remarkable adhesion to dentin. It seems to be suitable for use in periodontal tissues for retrograde root sealing and treatment of vertically fractured roots. For an adhesive resin to be useful clinically, it must bond not only to the dentin but also to the cementum.

The purpose of this study was to evaluate the long-term adhesion durability of 4-META/MMA-TBB resin to cementum kept in water at 37℃. Bovine root cementum and dentin surfaces were treated with 10-3 solution before 4-META/MMA-TBB resin was applied on treated surfaces. The micro-tensile bond strength (μTBS) decreased during the first two months; dye leakage value increased during the first four months and stabilized thereafter. No significant differences in adhesion durability were recognized between the dentin and cementum.

Key words : Adhesion, Durability, 4-META/MMA-TBB resin

INTRODUCTION

A number of retrograde filling materials have been tested for sealing the root canal. These materials include amalgam, reinforced zinc oxide-eugenol cement, and glass-ionomer cement. Clinical results in using amalgam is reported to have a 58.7-86.4% success rate[1-6], while that of Super-EBA™ is 65.4-86.4%[1,3,6]. Chem-Fil®[4] is reported to have an 87.8% success rate.

These clinical results are affected by many factors such as preoperative and operative conditions and methods. As to why they do not achieve good healing, one suggested reason is that the sealing capacity of the retrograde filling materials is insufficient. After using amalgam, reinforced zinc-oxide eugenol cement, and glass-ionomer cement for root-end filling, in vitro studies on dye leakage were carried out. It was found that, if it were over a long duration, application of these materials did not completely inhibit dye leakage[7-14].

Various in vitro studies have reported that good apical sealing was achieved by resin[15-17]. Vignaroli et al.[17] reported that applying bonding agents to the resected root-ends led to good apical sealing because of sealing of both the root canal and dentinal tubules. Not much research has been devoted to evaluating the clinical results of the use of adhesive resin for root-end sealing after apicoectomy. Only Rud et al. reported good results using Gluma and Retroplast™[18-21]. However, glutaraldehyde is included in Gluma, so there is concern about the possibility of harmful effects on the human body. Furthermore, contamination with blood of the resection surface during the application procedure causes inferior bonding and a risk of resin loosening. Resin is known to be sensitive to moisture during application. Therefore poor clinical results will arise when hemostasis is difficult to achieve intraoperatively, especially when failures in conjunction with poor hemostatic control during treatment have been evident[18].

4-META/MMA-TBB resin has been widely used in orthopedic and prosthetic dentistry. It has shown remarkable adhesive properties to dentin in vitro and in vivo[22-24]. Other favorable properties include little cytotoxicity, high level polymerization under wet conditions, and high biocompatibility after complete curing[25-27]. It was biocompatible and the sealing ability unaffected by contamination with blood for about five seconds after application[27]. So it seems to be also suitable for use in periodontal tissues. Thus it has been used for retrograde root sealing following apicoectomy or intentional replantation[28,29], as well as the bonding treatment of vertically fractured roots[30-32].

Clinical studies have suggested that 4-META/MMA-TBB resin might seal vertically fractured roots and prevent re-fractures in the bonding treatment[30-32]. It was reported that 4-META/MMA-TBB resin used as a root-end sealant following apicoectomy and intentional replantation induced a good prognosis at six months after surgery[28,29]. But an essential factor has not been looked into: the long-term adhesion between cementum and 4-META/MMA-TBB resin.

To be a suitable material in the bonding treatments of vertically fractured roots and root-end sealing, the adhesive resin must bond not only to dentin but also to cementum. Furthermore there should be

minimal leakage between the bonding material and the cementum, and the dentin as well. However, it has been demonstrated that the adhesive property of 4-META/MMA-TBB resin decreases when kept in water for long periods[23]. Not much research has been done to clarify the long-term stability of its adhesive bond strength and sealing property in the water.

The purpose of this study was to evaluate the adhesion durability of 4-META/MMA-TBB resin to cementum and dentin by employing the micro-tensile bond strength (μTBS) test and the dye leakage test using bovine teeth.

MATERIALS AND METHODS

Adhesive and bonding procedures
Ninety-six extracted bovine incisors were used in this study. Forty-eight teeth were used for the micro-tensile bond strength test, the rest used for the dye leakage test. After soft tissue and debris were removed from the tooth surface with a curette type scaler, the crowns were cut off at the cement-enamel junction with a low-speed diamond saw (Isomet, Buehler Ltd., Lake Bluff, IL, USA). Then the root pulp was extirpated with a barbed broach. Dentin specimens were obtained by removing cementum.

10-3 solution (activator Green®, SunMedical Co. Ltd., Shiga, Japan) was applied to the surface of the cementum with a Benda® Brush (Centrix, Shelton, CT, USA) for 5 seconds. The dentin surface was also treated with the same 10-3 solution for 5 seconds. Then 4-META/MMA-TBB resin (Super Bond C&B®, clear type, SunMedical Co. Ltd., Shiga, Japan) was applied on the treated surfaces according to the manufacturer's instructions using a brush dip technique. The diameter of the resin was approximately 3 mm. Three resin-dentin or resin-cementum bonded specimens were obtained per tooth. After resin dipping, all specimens were stored at 37℃ for 1 day, 1, 2, 3, 4 or 6 months. All samples were preserved in a solution of 10% Penicillin-Streptomycin (GIBCO™, Grand Island, NY, USA) and 10% AMPHOTERICIN B (Cellgro®, Herndon, VA, USA). The solution was changed once a week to prevent bacterial growth.

Micro-tensile bond testing
The specimens were sectioned perpendicular to the adhesive interface with a low-speed diamond saw under a water coolant. The sectioned specimens were carefully trimmed into an hourglass shape using a high-speed super-fine diamond point (SF 1104R, Shofu Co. Ltd., Kyoto, Japan) to form a gentle curve along the bonded interface from both sides to about 1.0 mm, thus resulting in an interface area of approximately 1 mm^2. These specimens were then attached to a testing apparatus with a cyanoacrylate adhesive (Model Repair II Pink, Dentsply-Sankin Industry Co. Ltd., Ohtawara, Japan) and subjected to micro-tensile bond testing[33-40] in a tabletop material tester (EZ Test, Shimazu Co., Kyoto, Japan) with a cross-head speed of 1.0 mm/min.

The μTBS values were expressed in MPa, and derived from dividing the imposed force (N) at the time fractured by the bond area (mm^2). When specimens failed before the actual testing, μTBS was determined from specimens that had survived the specimen processing with an explicit note of the number of pre-testing failures.

The bond strength data were analyzed by Mann-Whitney U test at 5% level of significance.

Fractographic analysis
After the micro-tensile bond testing was completed, all fractured surfaces were observed by a light microscope (OLYMPUS BX50, Olympus Ltd., Tokyo, Japan). Failure modes of resin-dentin and resin-cementum bonds were categorized into four types: cohesive failure of resin (R), cohesive failure of tooth (T), adhesive failure (A), and mixed failure (M). Fractographic analyses were statistically analyzed by the χ^2 test.

Dye leakage assessment
The specimens were transferred from the storage water into a 0.5% basic fuchsine solution and kept at room temperature for 24 hours. After being rinsed with water and air-dried, a section was made at the center of the material perpendicular to the bonding interface. The length of dye penetration and that of the interface between resin and tooth were measured with a measuring ocular in the light microscope. The maximum length was recorded for each specimen.

The leakage value was calculated as follows:

$$(\text{length ratio: } \%) = (\text{dye penetration length}/ \text{interface length}) \times 100$$

The leakage values were then analyzed by Mann-Whitney U test at 5% level of significance.

RESULTS

For dentin, μTBS decreased significantly from 28.4 MPa to 17.1 MPa in the first month (p<0.05), then remained stable at about 16 to 18 MPa from the first month to the sixth month. For cementum, μTBS decreased significantly from 32.9 MPa to 18.0 MPa over the first two months (p<0.05), then remained stable at about 18 to 20 MPa from month 2 to month 6. The μTBS differences between dentin and cementum were statistically significant (p<0.05) only at the first month, and no statistically significant differences were observed between month 2 and month 6 (Fig. 1).
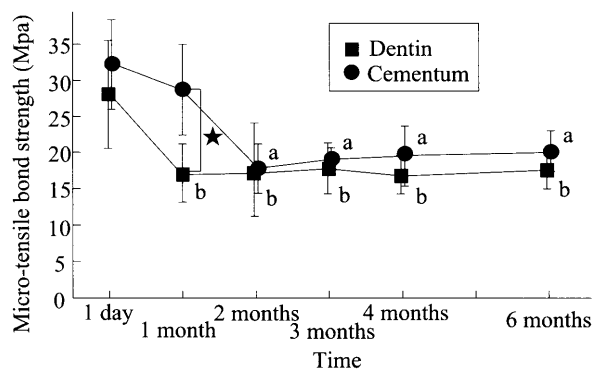
Fig. 1 Micro-tensile bond strength (mean ± standard deviation) to dentin and cementum by period after bonding. a and b indicates significant differences between day 1 and each time points in dentin and cementum (p<0.05). A star indicates a significant difference between dentin and cementum at one time point.

(a) Fracture modes of cementum specimens (%)

(b) Fracture modes of dentin specimens (%)

| R | cohesive failure of resin | T | cohesive failure of teeth |
| A | adhesive failure | M | mixed failure |

Fig. 2 Percentages of fracture modes of de-bonded surface. Within parentheses are the number of specimens that survived the preparation process per total number of specimens. (a) fracture modes of dentin specimens. (b) fracture modes of cementum specimens.
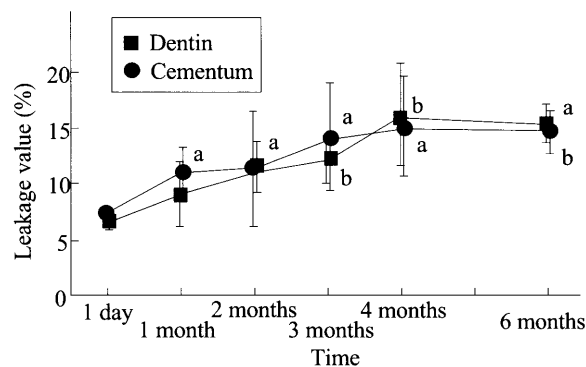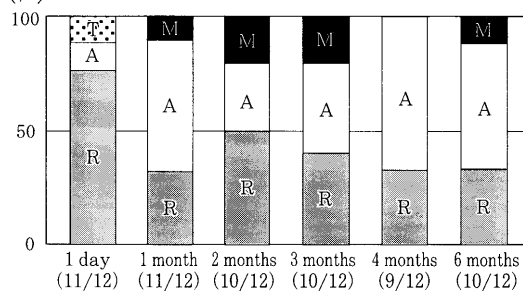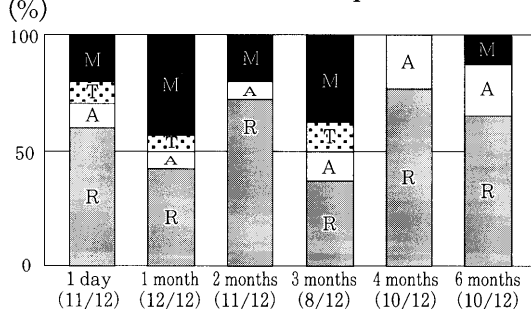

Fig. 3 Dye leakage value (mean ± standard deviation) of dentin and cementum by period after bonding. a and b indicates significant differences between day 1 and each time points in dentin and cementum.

According to fractographical analysis, the proportion of interfacial failure of the dentin sample tended to increase from 12.5% on the first day to 55.6% at sixth month. In the cementum sample, the destruction zone was not specified. No statistically significant differences were observed among all the fracture types (Fig. 2).

The leakage value of dentin increased significantly from 5.65% to 12.6% in the first three months (p<0.05); while for cementum, the leakage value increased significantly from 5.69% to 11.4% within the first month. For both dentin and cementum, no significant differences could be recognized at months 3, 4, and 6. The leakage values at sixth month for both dentin and cementum were almost the same (Fig. 3).

DISCUSSION

After day 1 in the micro-tensile bond strength test, the adhesive bond strengths of both dentin and cementum began to decrease. By the end of the first month, the adhesive bond strength of dentin had decreased by about 50%; for cementum, the strength decreased by about 50% at the second month. Following which there were no significant differences, and μTBS was maintained until the end of the six-month period. In the work of Kiyomura et al.[22], it was reported that the tensile strength decreased from 19.5 MPa to 7.9 MPa by the third month after the dentin samples were immersed in water. Kiyomura et al.[22] also reported that the samples' surfaces were treated with 10-3 solution for 30 seconds. It was recommended that 10-3 solution treatment be shorter than 30 seconds in order to expedite the generation of a long-term, stable hybrid layer. Therefore in this present study, treatment time with 10-3 solution was set at 5 seconds. The result was that the tensile bond strength to dentin decreased from 28.4 MPa to

17.6 MPa in three months. This result may lend support to the view that durable adhesive strength was achieved because the 5-second treatment time seemed to prevent over-etching of the dentin.

It was suggested that due to water immersion for long periods, failure occurred between the hybrid layer and the resin layer, or between the hybrid layer and the mineralized dentin[23]. In several studies, bond strength was reduced after resin-dentin bonded specimens were stored long-term in water[22,41,42]. It has been speculated that water absorbed into the demineralized dentin had caused hydrolytic degradation[22,23,41,42] of the resin, thus leading to non-enveloped collagen fibrils at demineralized dentin zone. Hence, the presence of demineralized dentin was thought to have affected the integrity of the resin-dentin bonds for both short and long periods[44]. Several studies concluded that a prolonged acid conditioning time resulted in fracture within the demineralized dentin zone when specimens were stressed to failure using the micro-tensile bond test[43,44]. This means that excessive acid conditioning caused deeper demineralization of both intertubular and peritubular dentin, which in turn meant that they were not capable of being completely infiltrated by resin monomers. These studies suggested that failure was likely to be initiated in this weakest zone, leading to decreased bond strength[45]. In this study, μTBS decreased at the initial stage with no significant differences afterwards. The reason for this change is unclear, but Kiyomura et al. supported the view that bonding strength decreases gradually following the initial decline of adhesive strength[22].

On fractographic analysis, singularity was not recognized in either the dentin sample or the cementum's. The degradation effect on adhesion due to long-period water immersion has not been established. But in dentin, it was recognized that the adhesive interface tended to degrade as time progressed. So demineralized dentin was deemed to be incapable of being completely infiltrated by resin monomers. It was reported that long-term morphological changes had occurred in both the adhesive interface and the resin-composite restorative materials[46,47]. In the present study, there were no significant differences between the adhesive and cohesive failure rates of resin. Hence, a dominant factor for failure was lacking. In cementum however, there tended to be a much higher rate of cohesive fracture, though the results were not statistically significant. Adhesive failure rate, on the other hand, seemed to be lower when cementum surfaces were treated with 10-3 solution for 5 seconds. The μTBS of resin-cementum at first month was significantly higher than that of resin-dentin, and the rapidity of dentin's adhesive strength decline was clearly significant. Two reasons were suggested for the vast difference. First, owing to the greater density of cementum (due to the absence of

tubules), water can infiltrate the dentin more easily. Secondly, it could be that demineralized cementum was capable of being completely infiltrated by resin monomers.

The leakage values of dentin and cementum increased until the third month and then remained fairly constant until the sixth month. At sixth month, the leakage value was about 15% in the 3-mm-diameter specimens. In the dye leakage test, leakage is identified by the permeation of dye molecules in the gap between resin and dentin. The gap could be formed due to curing shrinkage of the resin or degeneration of the adhesive interface. It is known that polymerization continues for 4-META/MMA-TBB resin for about one month[48,49]. The continuance of polymerization expanded the contraction gap, hence the leakage quantity increased. However, the gap did not seem to expand during the last two months of the experiment — as indicated by the stabilized leakage values.

Through the micro-tensile and dye leakage tests, this study has shown that for both dentin-resin and cementum-resin bonds, the declining tendency of adhesive strength was not significant after the first 2-3 months. This means that it is highly likely that adhesion would remain strong and durable for a long term. Based on these test results, it seems promising that 4-META/MMA-TBB resin would be clinically effective for the bonding treatments of vertically fractured roots and root-end sealing following apicoectomy, in which bonding to cementum is necessary.

CONCLUSION

From the results of micro-tensile and dye leakage tests, it became clear that the adhesion between 4-META/MMA-TBB resin and dentin or cement decreased for 1-3 months, and then stabilized afterwards. During most of the observation periods, no significant differences were recognized between dentin and cementum.

ACKNOWLEDGEMENTS

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