Evaluation of a New Adhesive Liner as an Adhesive Promotor and a Desensitizer on Hypersensitive Dentin

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The effect of N-methacryloyl aminosalicylic acid (NMSA) on the tensile bond strength between resin materials and bovine dentin and also its desensitizing efficacy on hypersensitive dentin were investigated.

In case of a composite resin, significantly greater bond was obtained by a combined treatment with 10 % citric acid containing 3 % CuCl_2 (10-3 solution) and NMSA-liner, than that of the control group treated by H_3PO_4 only. Bond strength of MMA/TBB system resin was also promoted by NMSA application. In the clinical evaluation, the application of NMSA-liner has relieved most cases of the dentin hypersensitivity immediately after even a simple coating. For obtaining the long term effect, the dentin surface should be treated at first by 10-3 solution, though the efficacy was eliminated after a few days in some cases. Accordingly, this material seems to be promising both in the field of conservative dentistry and adhesive dentistry.

Key words : NMSA, Adhesion promotor, Desensitizer

INTRODUCTION

The development in a adhesive dentistry has rapidly grown in the past few years by the improvements of adhesive resin materials which bond strongly, particularly to dentin¹⁻³⁾. It is generally considered, however, that the composite resin restorations with the acid-etch procedure irritate the dental pulp^{4,5)}. On the other hand, some researchers have proved that a good bond and adaptation between the resin restoration and dentinal walls did not cause significant pulpal responses⁶⁻⁸⁾. However, we, in the routine clinic situation, are still bothered by postoperative sensitivity. Recently, a bonding liner containing an aromatic monocarboxylic acid derivative, which was expected to promote the adhesion between a resin material and enamel, has been reported to be effective even for protection against dental pulp irritation⁹⁾.

From another approach we predicted a more probable desensitizing effect of a salicylic acid derivative on dentin. Fortunately, Kojima *et al.*¹⁰⁾ have reported the copolymerization of N-methacryloyl aminobenzoic acids and aminosalicylic acids with MMA. Additionally, 2 % MMA solution of these acids were prepared in adhesion between ivory and acrylic resins, and N-methacryloyl 5-aminosalicylic acid produced the strongest adhesion, showing approximately 20 MPa. The purpose of this study was to evaluate the effects of this material when modified as an adhesive liner, not only as an adhesion promotor, but also as a desensitizer on hypersensitive dentin.

MATERIALS AND METHODS

1. Tensile adhesion test

Bovine teeth, kept in a refrigerator, were used. For the adherend, flat dentin surfaces were prepared by grinding buccal enamel and polishing it with a #600 waterproof silicon carbide paper.

The dentin surfaces were conditioned for 1 minute each with the following acidic solutions: 37 % phosphoric acid (H_3PO_4) and 10 % citric acid containing 3 % cupric chloride (10–3 solution).

After washing and drying the dentin surfaces, 5 % NMSA (N-methacryloyl 5-aminosalicylic acid, Fig. 1) in acetone solution (NMSA-liner) was used as an adhesive liner. NMSA was prepared by the reaction of methacrylic chloride with 5-aminosalicylic acid. 4META (4-methacryloxyethyl trimellitic anhydride) was also made by the condensation of 2-hydroxyethyl methacrylate and trimellitic anhydride acid chloride, with pyridine as the hydrochloric acid acceptor. Five percent 4META in acetone solution (4META-liner) was employed as a control liner.

Fig. 1 N-methacryloyl 5-aminosalicylic acid.

An attached bonding agent of a composite resin* was thinly coated on the lined dentin, before the composite resin was inserted in the copper tube placed on the dentin surface. The testing method was the same as that described previously¹¹⁾.

In addition, the bond strength of the mixture of PMMA (polymethylmethacrylate) and MMA (methylmethacrylate) activated by TBB (tributylborane) to dentin was also examined, by carefully placing an acrylic rod on the lined dentin surfaces according to the method described before²⁾. MMA containing 2% NMSA was also tested without liner. As a control, Super Bond C & B** was used without liner because this product is composed of PMMA and MMA containing 5 % 4META activated by TBB.

Ten minutes after mixing the materials, the bonded specimens were immersed in water at 37°C and stored for 1 day before the tensile bond strength was determined by the Instron-type universal testing machine***.

Ten specimens were used in each group.

2. Analysis of the desensitizing effect of NMSA on hypersensitive dentin

The desensitizing effect of NMSA-liner on hypersensitive dentin was clinically evaluated. This evaluation was conducted on the teeth of 76 healthy people, who had suffered from dental hypersensitivity accompanied by gingival recession or/and abrasion.

At first, the hypersensitive area was carefully tested by cold air stimulus, using an air gun

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

** Sun Medical Co., Tokyo, Japan

*** Shimadzu Autograph AG-500B, Shimadzu Co., Kyoto, Japan

attached to the dental chair and given a score as the baseline. The treatment with 10-3 solution was performed for 30 seconds to clean the surface of hypersensitive area. After washing and drying, NMSA-liner was applied with a sponge pellet. After the evaporation of acetone, the same hypersensitive area was retested by cold air for assessing the effect immediately after application. One week after the application, the treated area was checked again in the same way.

The NMSA-liner application without 10-3 treatment was also evaluated.

Rating scales for the responses to the stimulus were ranked as follows: 0, no pain; 1, awareness of stimulus or discomfort; 2, pain, but not severe; 3, severe pain. By the comparison of the rank between evaluations at the baseline and each period after the liner application, the desensitizing efficacy of the NMSA-liner was judged as follows: remarkable effect, which indicates decrease in score from 3 to 1 or 0 and from 2 to 0; moderate effect, which indicates decrease in score from 3 to 2, from 2 to 1 and from 1 to 0; none effect, which indicates no change at all.

3. SEM investigation

The dentin of the extracted human incisor, ground and polished with a #600 water-proof silicon carbide paper and treated variously with 10-3 solution and liners, was investigated by a SEM****.

RESULTS

1. Adhesion

Table 1 shows the tensile adhesion of the composite resin to bovine teeth after various treatments. These results were discussed with analysis of variance followed by Newman Keul's tests. The strongest bond was obtained from the treatments with 10-3 solution and 4META-liner. The control group treated by H_3PO_4 without any liner produced the weakest bond of 3.4 MPa, showing significant differences from those of the groups treated with 4META-liner or NMSA-liner.

Table 1 Tensile bond strengths of a composite resin bonded to treated bovine dentin

Surface treatment	Liner	Adhesion (S. D.) MPa
10-3	4META-liner	9.9 (3.4)
10-3	NMSA-liner	6.9 (1.7)
H_3PO_4	4META-liner	8.5 (1.8)
H_3PO_4	NMSA-liner	5.9 (2.0)
H_3PO_4	—	3.4 (1.3)

Vertical bars on the right indicate that the figures at both ends are significantly different ($p < 0.05$).

When the PMMA/MMA-TBB system resin was used, the bond strength was much stronger than that of the composite resin (Table 2). The groups treated with H_3PO_4 solution showed significantly smaller bond strength than that of the groups treated with 10-3 solution. Combined treatment with 10-3 solution and NMSA-liner produced very strong bonds, similar

**** JXA-840, JEOL Ltd., Tokyo, Japan

Table 2 Tensile bond strengths of PMMA/MMA-TBB resin bonded to treated dentin

Treatment	Liner	MMA Monomer	Adhesion (S. D.) MPa
10-3	NMSA-liner	MMA	17.3 (4.5)
10-3	—	2% NMSA/MMA	18.1 (2.7)
10-3	—	5% 4META/MMA	17.2 (3.7)
H ₃ PO ₄	NMSA-liner	MMA	7.9 (1.9)
H ₃ PO ₄	—	2% NMSA/MMA	13.5 (4.1)
H ₃ PO ₄	—	5% 4META/MMA	10.6 (2.4)

Vertical bars on the right indicate that the figures at both ends are significantly different ($p < 0.05$).

to that of Super Bond C&B. When MMA containing 2 % NMSA was used, a strong bond was also obtained, the same as with Super Bond C&B.

2. Desensitizing effect

Table 3 shows the results of the clinical evaluations. When NMSA-liner was applied after 10-3 treatment, 99 % rated effective immediately after application. That is, 87 % of the teeth showed remarkable effect, 12 % were moderate, and only 1 case showed no effect. But 1 week after the application, 29 % had no effect. A significant difference was found between the immediate postapplication evaluation and the evaluation after 1 week, according to a rank sum test (Wilcoxon's test). When the liner was applied without 10-3 treatment, 83 % of the teeth was rated effective at the immediate postapplication evaluation. But after 1 week, 55 % were not effected.

Table 3 Results of clinical evaluation

10-3	Evaluation Period	remarkable	moderate	none
treated	immediate	94 (87%)	13 (12%)	1 (1%)
	1 week	68 (63%)	9 (8%)	31 (29%)
untreated	immediate	22 (55%)	11 (28%)	7 (18%)
	1 week	7 (18%)	11 (28%)	22 (55%)

Significant differences ($p < 0.01$) were found among the all groups.

Fig. 2 Dentinal surface ground and treated with 10-3 solution. Dentinal tubules are opened with the 10-3 treatment.

Fig. 3 Dentinal surface applied NMSA-liner after grinding and 10-3 treatment. Dentinal tubules are still opened.

Fig. 4 Profile view of the specimen in Fig. 3. Dentinal tubules are not occluded even at the deep area.

When comparing 10-3 treated group with the untreated group, the significant differences were found in both evaluations immediately after application and 1 week after application.

3. SEM investigation

Figure 2 shows the dentin treated with 10-3 solution, indicating the tubules opened when the smear layer and the dentinal plug dissolved. The tubules were still open in spite of the application of NMSA-liner (Fig. 3). Even in the observation of the divided specimen, the occlusion at the deep area of the tubules could not be confirmed (Fig. 4).

DISCUSSION

In this study, the application of NMSA-liner increased the bond strength of a composite resin or MMA/TBB resin to bovine dentin. In the case of the composite resin, 4META-liner produced a stronger bond than NMSA-liner. The bond strength of the mixture of PMMA and MMA containing 2 % NMSA was similar to that of Super Bond C&B.

Bowen *et al.*¹¹ reported that applications of a cleanser/mordant solution, a surface active comonomer, and a coupling agent, produced a strong bond of composite resin to dentin. Though a mild acidic solution is recommended to remove the smear layer¹², dentin etching with H_3PO_4 solution has been performed to obtain a stronger bond of composite, in Japan^{11,13}. In this study, however, the treatment with H_3PO_4 was confirmed not to be suitable for bonding of MMA/TBB resin to dentin. The result coincides with a previous study by Nakabayashi *et al.*². The treatment with 10-3 solution is considered best to prepare a suitable condition for 4META or NMSA to be used on dentin.

Unfortunately, the etiology of the dentinal hypersensitivity has so far been unclear because the mechanism of pain transmission within the dentin is still doubtful. Although several theories have been suggested for hypersensitive dentin, it appears the hydrodynamic theory is most plausible. And the applications of medicaments aiming at the occluding of the dentinal tubules have generally been used in the clinic on a trial and error basis. However, Mosteller¹⁴ reported that predonisolone eliminated the postoperative thermal sensitivity of dentin. The authors anticipate some anodyne effect of NMSA, because it is an aminosalicylic acid derivative. In preliminary investigation, the dose-toxic effect of this material was confirmed to be similar to that of para-aminosalicylic acid (PAS) which is an antituberculosis drug, though the pharmacological effect of NMSA has not been investigated yet. Actually, the desensitizing effect was observed in a clinical trial, when used as the adhesive liner before the composite restoration. Thus, the preliminary clinical evaluation was performed.

On the other hand, it has been reported that the occlusion of the patent tubules established a protective diffusion barrier, and that the hydraulic conductance of dentin increased when etched with acid¹⁵. In this study, however, the application of NMSA-liner with 10-3 treatment was more effective when compared to the application without 10-3 treatment. The SEM investigation did not show the image of occlusion nor sealing of the opened tubules. Subsequently, it is considered that the patent tubules and the dentinal surfaces conditioned by 10-3 solution would promote the penetration of NMSA-liner. This also suggests that even in case of application of other desensitizing agents, the specific surface cleansing or treatment suitable for each medicament or agent may increase the effect.

The fact that almost all the cases showed some desensitizing effect immediately after the application of NMSA-liner with or without 10-3 treatment might also suggest that it allows for increased accommodation to sensitivity, as was seen by the repeated cold air stimulus during the procedure and the cooling of the tooth by the evaporation of acetone. Even if NMSA has an anodyne effect, the effect seems not to continue very long. The temporary changes of threshold pain might change the sensitivity of the tooth.

Though further investigation needs to be done, the application of this liner is considered to be very useful in the clinic, since it is effective not only for the hypersensitivity, but also for the bonding of composite resin. Furthermore, it may be able to decrease the postoperative sensitivity, contribute to good restoration for the root surface caries, and bring about great benefits in geriatric dentistry.

CONCLUSION

1. NMSA-liner produced a strong bond of composite resin or MMA/TBB resins to bovine dentin.
2. The use of 10-3 solution may provide a compatible condition for NMSA-liner to be used on dentin.
3. NMSA-liner was effective in desensitizing hypersensitive dentin, especially immediately after the application, but the effect diminished after 1 week in some cases.

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また軟組織との反応において中実体では Zr-Ti 合金はアパタイトやチタンとはあまり差異がみられなかつ

た。多孔材では上皮が多孔材部を完全におおうように治癒した場合に結合組織が入り強固な結合が得られた。

新しい接着性ライナーの接着促進効果と象牙質知覚過敏鈍麻作用の評価

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サリチル酸の誘導体である N-メタクリロイルアミノサリチル酸 (NMSA) に着目し, その接着性ライナーとしての評価のみならず象牙質に対する知覚鈍麻作用についても検討した。

その結果, 5%NMSA アセトン溶液を調整し NMSA ライナーとして用いると, 牛象牙質に対する各種レジン材料の引っ張り接着強さが向上することが確認された。

また象牙質知覚過敏症に対する本剤の塗布効果を臨床的に評価したところ, 塗布直後においては, ほとんどすべての症例において症状の消退がみられ, 臨床的に極めて効果的であることが示唆された。数日後にその効果の消退する例もみられたが, 塗布面に対する 10-3 溶液による前処理の有効性も確認された。本剤の象牙質知覚過敏症に対する作用機序については現在検討中である。