Efficacy of a self-etching dentin primer composed of TEGMA and phenyl-P

Shinichiro Yoshimoto* / Kazuo Itoh** / Atsufumi Manabe*** / Mitsuko Inoue**** / Hisashi Hisamitsu***** / Ryuuji Sasa******

The purpose of the present study was to evaluate the efficacy of an experimental self-etching dentin primer composed of TEGMA and phenyl-P using primary and young permanent teeth. The efficacy of the self-etching dentin primer was evaluated by measuring the wall-to-wall polymerization contraction gap width and the shear bond strength to the flat dentin surface. The contraction gap formation was prevented completely in the specimens primed with the 35 vol% TEGMA and 20% phenyl-P for 30 sec.
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INTRODUCTION

Since the report of GLUMA primer by Munksgaard and Asmussen in 1984[1], dentin priming has been recognized as a mandatory step to establish the dentin bonding. They defied that the bond strength of the resin monomer to the EDTA-conditioned flat dentin surface was significantly improved by pretreatment with aqueous mixture of glutaraldehyde and 2-hydorxyethyl methacrylate (2-HEMA). In 1985[2-5] we reported that the glutaraldehyde was not essential because the 2-HEMA solution exhibited a comparable priming effect to that of the GLUMA primer if the commercial dentin bonding agent, which contained 10-methacryloxydecyl dihydrogen phosphate (10-MDP)[6] was applied after priming. The dentin primer should be consistently applied to the dentin after removing the smear layer by using the dentin conditioner. However, due primarily to the complexity of the clinical handling of the dentin bonding system, the self-etching dentin primer was commercially introduced in 1988.[7-9] Most of the commercially available self-etching dentin primers were composed of 2-HEMA solution and either an acid or an acidic monomer, such as maleic acid,[10] succinic acid or the functional monomer of 10-MDP, to dissolve the smear layer. In 1990 and 1992[11,12] Watanabe *et al.* reported that the aqueous mixture of 2-HEMA and methacryloxyethyl hydrogen phenyl-phosphate (phenyl-P) exhibited remarkable efficacy as a self-etching dentin primer. In addition, Chigira *et al.*[15] reported that the experimental self-etching dentin primer composed of an aqueous mixture of phenyl-P and 2-HEMA or glyceryl mono-methacrylate (GM) completely prevented the contraction gap formation of the light activated resin composite in a cylindrical dentin cavity.

As reported by Hayakawa *et al.* and others,[16-21] clinical use of 2-HEMA solution is quite doubtful because the solution causes contact dermatitis as a delayed allergic reaction. After the first report of the GLUMA primer, dentin primers composed of polyvalent alcohol that was esterified with methacrylate, glyceryl mono-methacrylate (GM),[22] erythritol mono-methacrylate[23] and xylitol mono-methacrylate[24] were developed. Furthermore, two diol solutions, ethylene glycol and 1,6-hexane[25] diol were reported as contraction gap-free dentin primers. All of these experimental dentin primers were reported to avoid the side effects associated with the 2-HEMA solution. In 2002, we reported that both triethylene glycol (TEG) and triethylene glycol mono-methacrylate (TEGMA) solutions prevented the contraction gap formation as the dentin primer. The purpose of the present study was to evaluate the efficacy of an experimental self-etching dentin primer composed of the TEGMA and phenyl-P.

* Shinichiro Yoshimoto, D.D.S, Assistant, Showa University School of Dentistry, Tokyo, Japan.
** Kazuo Itoh, D.D.S, D.D.Sc, Associate Professor, Showa University School of Dentistry, Tokyo, Japan.
*** Atsufumi Manabe, D.D.S, PhD, Assistant Professor, Showa University School of Dentistry, Tokyo, Japan.
**** Mitsuko Inoue, D.D.S, D.D.Sc, Associate Professor, Showa University School of Dentistry, Tokyo, Japan.
***** Hisashi Hisamitsu, D.D.S, D.D.Sc, Professor, Showa University School of Dentistry, Tokyo, Japan.
****** Ryuuji Sasa, D.D.S, D.D.Sc, Chairman and Professor, Showa University School of Dentistry, Tokyo, Japan.

Send all correspondence to Dr. Atsufumi Manabe, Department of Operative Dentistry, Showa University School of Dentistry, 2-1-1 Kitasenzoku, Ohta-ku, Tokyo 145-8515, Japan.

Voice: +81-3-3787-1151
Fax: +81-3-3787-1229
E-mail: amanabe @ senzoku.showa-u.ac.jp

MATERIALS AND METHODS

The experimental self etching dentin primer was prepared as an aqueous mixture of 35 vol% tri-ethyleneglycol mono-methacrylate (TEGMA) and methacryloxyethyl hydrogen phenyl phosphate (phenyl-P) at concentrations ranging from 5 to 20 vol%.

Extracted primary and young permanent human teeth were used in the present study.

The efficacy of the self-etching dentin primer was evaluated by measuring the wall-to-wall polymerization contraction gap width of the light activated resin composite in the cylindrical dentin cavity and the shear bond strength to the flat dentin surface.

The proximal enamel of the extracted human teeth was eliminated, producing a flat surface, and a cylindrical cavity, 3mm in diameter and 1.5mm in depth was prepared in the exposed dentin. The self-etching dentin primer was applied in the cavity for 10 to 30 sec followed by air blasting. Then, a commercial dual-cured dentin bonding agent containing 10-MDP (Clearfil Photo Bond, Kuraray, Okayama, Japan) was applied in the cavity and was irradiated for 10 sec after eliminating the excess materials by gentle air blast. The light activated resin composite (Palfique Estelite, Tokuyama, Yakaguchi, Japan) was slightly over-filled. The composite surface was gently pressed on a glass plate, momentarily mediated with a plastic matrix and then irradiated for 40 sec. After storing the specimens in water at room temperature (24 ± 1˚C) for 10 min, the cavity marginal was exposed on a wet carborundum paper and polished on a linen cloth mediated with alumina slurry having a grain size of 0.03μm. The marginal adaptation of the resin composite was inspected via light microscope and the possible gap width was measured at eight points every 45 degrees along the cavity margin. The contraction gap value was determined as the sum of the diametrically opposing gap widths as a percentage of the cavity diameter. The maximum of four gap values was given as the contraction gap value of the specimen. Ten specimens for each component and priming time, in total, were prepared.

The extracted human tooth was embedded in an epoxy resin and the flat dentin surface was prepared using a wet carborundum paper grit number of 220. The split Teflon mold inner diameter of 3.6mm, outer diameter of 20mm and 5.0mm in height was clamped on a specimen. The dentin priming and the dentin bonding agent application was performed using the same method as described above and the resin composite was placed in the Teflon mold to a thickness of not more than 3mm. The resin composite was irradiated for 40sec from the top of center hole of the mold. Ten minutes after the irradiation, the mold was removed and the shear bond strength was measured using the universal testing machine.

Negative control specimens were created by: 60-second 35 vol% TEGMA application - drying - Clearfil Photo Bond application - 10-second beam irradiation - Estelite filling - 40-second beam irradiation.

Positive control specimens were created by: 60-second EDTA application - washing and drying - 60-second 35 vol% TEGMA application - drying - Clearfil Photo Bond application - 10-second beam irradiation - Estelite filling - 40-second beam irradiation.

RESULTS

The contraction gap formation of the resin composite was prevented completely in the specimens that were primed with the aqueous mixture of 35 vol% TEGMA and 20% phenyl-P for 30 sec (Tables 1-a and 1-b). However, complete marginal integrity was not obtained when either the concentration of the phenyl-P or the priming time was reduced. The mean shear bond strength of the tested bonding system varied from 5.163±3.574 to 11.230±1.699 Mpa and significant differences were observed between priming using 10-35PP-TEGMA for 20 sec and 20-35PP-TEGMA for 20 sec. Although the value of 20-35PP-TEGMA for 30 sec was very low, a good contraction gap value was obtained.

The positive control specimen exhibited significantly higher bond strengths than all experimental groups (Tables 2-a and 2-b). Furthermore, there was no significant difference observed between the marginal adaptation of primary teeth and permanent teeth.

When 20-35PP-TEGMA was applied to primary teeth for 30 sec, all the contraction gaps were 0%, indicating complete adaptation to the cavities. The contraction gaps were also all 0% for the positive control on primary teeth. The shear bond strengths of the primary teeth were 9.282±3.033 Mpa (Tables 3-a and 3-b).

Therefore, the 30-second application of 20-35PP-TEGMA showed equal cavity adaptation between primary teeth and permanent teeth. In other words, the experimental self-etching dentin primer was effective for the dentin of both primary teeth and permanent teeth.

DISCUSSION

Self-etching dentin primers have been introduced in clinics to simplify the procedures for dentin bonding by combining the steps of dentin conditioning and priming. In the case of pediatric dentistry, the experimental self-etching primer was very useful as a filling resin composite. This study suggests that the experimental primer has both high marginal adaptation and high safety with respect to oral tissue. The smear layer on the ground tooth surface should be removed because this layer disturbs the bonding between the tooth and the dental adhesives. Although various acids and acidic monomers in commercially available materials were introduced, no consistent conclusion has been reported concerning the type of acid in the dentin conditioner and the self-etching dentin primer. Most of the commercial self-etching dentin primers are composed of 2-HEMA solution and either an acid or an acidic monomer, because their components have only modified of those of the GLUMA primer. The clinical use of 2-HEMA solution has been strongly advised against

Table 1-a. Contraction gap width with permanent teeth

Application Time	5-35PP-TEGMA	10-35PP-TEGMA	20-35PP-TEGMA
10 sec	0.057±0.054(3) I	0.014±0.023(7) I	0.029±0.049(7) I
20 sec	0.019±0.038(7) I	0.016±0.034(8) I	0.012±0.038(9) I
30 sec	0.025±0.042(7) I	0.015±0.046(9) I	0 (10) I

%, N=10 Mean±SD, Number of gap-free specimens ( )
I : Significantly different based on the results of a Kruskal-Wallis test ($p<0.05$).

Table 1-b. Contraction gap width with permanent teeth

	negative control	positive control
	0.083±0.076(2)	0 (10)

%, N=10 Mean±SD, Number of gap-free specimens ( )

Table 2-a. Shear bond strength with permanent teeth

Application Time	5-35PP-TEGMA	10-35PP-TEGMA	20-35PP-TEGMA
10 sec	7.757±4.334 I	6.255±1.355 I	9.387±3.222 I
20 sec	7.847±3.893 I	5.163±3.574 I	11.230±1.699 I
30 sec	7.239±1.676 I	8.251±0.338 I	6.085±1.761 I

Mpa, N=5 Mean±SD
I : Significantly different based on the results of a Kruskal-Wallis test ($p<0.05$).

Table 2-b. Shear bond strength with permanent teeth

	negative control	positive control
	6.943±3.221	24.161±5.162

Mpa, N=5 Mean±SD

Table 3-a. Contraction gap width with primary teeth

Application Time	20-35PP-TEGMA	negative contro	positive control
30 sec	0 (10)	0.124±0.074(2)	0 (10)

%, N=10 Mean±SD, Number of gap-free specimens ( )

Table 3-b. Shear bond strength with primary teeth

Application Time	20-35PP-TEGMA
30 sec	9.282±3.033

Mpa, N=5 Mean±SD

because severe side effects on the soft tissue may occur. We reported the experimental contraction gap-free dentin primers, diols or polyvalent alcohols esterified with methacrylate.[22-25] In addition, triethyleneglycol (TEG) and triethyleneglycol mono-methacrylate (TEGMA) solutions were recently developed to avoid the side effects of the 2-HEMA primer. Polyethylene glycol (PEG)[26,27] has been used as a hydro-gel capsule in medicine to prolong metabolism time, resulting decreased frequency of drug injection. This effect has been dubbed "pegylation". The PEG tested in this study had a relatively low molecular weight and was esterified with methacrylate. The optimum concentration and molecular weight of the PEG when used as a dentin primer should be examined in greater detail.

Self-etching dentin primers have been widely used even though the detailed mechanism thereof has not been clarified completely. The acid or acidic monomer in the primer is essential for dissolving the smear layer. Therefore, most self-etching dentin primers should be applied in the cavity for not less than 30sec. The priming effect of commercial materials containing 2-HEMA solution has been explained by either the promotion of the adhesive monomer diffusion into the dentin or the expansion of the collagen network that was exposed by dentin etching and then collapsed by air blasting.[28-34] However, these explanations are conflicting because the dentin bonding agent bond strength to the enamel is significantly stronger than that to the dentin. In most of the above mentioned reports, the dentin bonding agent is considered to bond to the inorganic component in the enamel and to the organic component in the dentin in the same cavity. Adhesive monomers such as 4-methacryloxyethyl trimellitate anhydride (4-META)[35-37] and 10-MDP have a chemical structure that allows bonding to inorganic substances, but not to inorganic substances. We demonstrated that the contraction gap width of the resin composite in the cylindrical dentin cavity was increased when the cavity wall was decalcified by dentin conditioner and when the functional monomer in the dentin bonding agent was eliminated. Therefore, it is believed that the dentin bonding was established by the interaction between the inorganic component in the dentin structure and the functional monomer in the dentin bonding agent.[38]

The primary requirement for the dentin bonding system is to prevent the separation of the resin composite paste from the cavity wall during polymerization shrinkage. We claimed that the efficacy of the dentin adhesives should be evaluated by contraction gap observation in the three-dimensional cavity and not by bond strength measurement of the two-dimensional flat tooth surfaces. The interaction between the efficacy of dentin adhesives and the polymerization contraction stress of the resin composite can not be detected by the bond strength measurement. Organic attack dentin bonding theories are based on either bond strength measurement or microstructure observation at the adhesive interface prepared by the dentin adhesive sandwiched between two dentin discs. Observation of the microstructure is performed in a high vacuum chamber, which is extremely different from the oral environment.

Firm bonding between the acid-etched enamel and the dentin bonding agent was obtained. The priming procedure is required only for the dentin cavity wall, probably because the physical characteristics of the dentin might be made to resemble that of the enamel. As demonstrated by Ohhashi[39] and Kusunoki[40] the contraction gap formation was prevented completely by the momentary priming using 35vol% of GM, TEG and TEGMA solution. The priming by GM, TEG or TEGMA solution is thought to instantly alternate the physical characteristics on the dentin, resulting in improved bonding efficacy. The physical characteristics of the dentin might be improved for bonding resemble to that of the enamel. Thus, the dentin bonding agent bonds both to the enamel and to the dentin by the same mechanism, although the details of this mechanism should be investigated.

The phenyl-P tested in the present study was primarily employed as the adhesive monomer in commercial dentin bonding agents in 1979.[41] Watanabe *et al.*[11,12] reported that the aqueous mixture of phenyl-P and 2-HEMA exhibited a significantly higher efficacy as the self-etching dentin primer than did the 4-META or the metacryloxyethyl phosphate in the 2-HEMA. As demonstrated in the present study, the high efficacy of phenyl-P as the acidic monomer in the self-etching dentin primer was ensured. Further study is necessary in order to define the optimum molecular weight of the polyethylene glycol or the polyethyleneglycol monomethacrylate for the self etching dentin primer.

The use of the experimental self-etching dentin primer successfully reduced the number of treatment steps as well as the processing time down to 30 seconds. This primer is considered to be effective for repairing composite resin in treating the teeth of children, where the primary teeth are small, making it difficult to distinguish the enamel and the dentin.

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