2-HEMA-free Dentin Bonding System to Prevent Contraction Gap

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The effects of 2-hydroxyethyl methacrylate (2-HEMA) both in the primer and in the commercial dentin bonding agent on the efficacy of the dentin bonding system was evaluated by measuring the polymerization contraction gap width of a commercial resin composite restored in a cylindrical dentin cavity prepared in an extracted human molar. Contraction gap formation was prevented in the group of the EDTA-conditioning followed by glyceryl mono-methacrylate, ethylene glycol and 1,6-hexanediol priming regardless of the 2-HEMA content in the dentin bonding agent containing 10-methacryloxydecyl dihydrogen phosphate. However, gap formation could not be prevented completely by the 2-HEMA priming. Therefore, it was possible to conclude that 2-HEMA was neither essential in the primer nor in the dentin bonding agent because of its low bonding efficacy and the resulting side effects on skin tissue as a delayed allergic reaction.

Key words: 2-HEMA, Contraction gap, Dentin bonding

INTRODUCTION

2-hydroxyethyl methacrylate (2-HEMA) has been widely used as one of the main components of the primer bonding agent in dentin bonding systems. The efficacy of 2-HEMA solution as a dentin primer was initially introduced by Munksgaard and Asmussen. They suggested that dentin collagen was activated by the glutaraldehyde and consequently polymerized with 2-HEMA. The priming effect of the 2-HEMA solution and other methacrylate derivatives was recently explained by expanding the collagen network in which the resinous dentin adhesives impregnated and polymerized, or by promoting bonding monomer diffusion into the profound dentin layer which was desirable to form a hybrid layer.

However, the undesirable side effect of the 2-HEMA monomer against the skin tissue and oral mucous was strongly attacked by Kanerva et al., Hayakawa et al. and Katsuno et al. They reported that the 2-HEMA solution caused contact dermatitis as a delayed allergic reaction after direct contact with the 2-HEMA solution.

To avoid such side effects, Chigira et al., Manabe et al. and Yokoi et al. studied glyceryl mono-methacrylate (GM), meso-erythritol mono-methacrylate (EM) and xylitol mono-methacrylate (XM) solutions as alternative dentin primers, each of which exhibited significantly higher efficacy than the 2-HEMA primer. Contraction
gap formation in a commercial light-activated resin composite was prevented completely when primed using an aqueous solution of these methacrylate derivatives, whereas it was impossible with 2-HEMA priming\(^{17}\). However, the side effects of these solutions would not be neglected completely because of the cross effect among the 2-HEMA, GM, EM and XM solutions\(^{16}\).

Ohhashi \textit{et al.}\(^{19}\) and Rahman \textit{et al.}\(^{20}\) developed contraction gap-free dentin primers composed of diol solution, ethylene glycol (EG) and 1,6-hexanediol (HD), based on the speculation that the allergic reaction might be due to the methacrylate group of these methacrylate derivatives. This finding suggested that the polymerization group was not mandatory to exhibiting high efficacy as a dentin primer. Therefore, the clinical use of the 2-HEMA primer has not been recommendable because of its side effects on soft tissue and its low priming effect compared with the GM, EM and XM solutions and two-diol solutions.

Even when 2-HEMA was eliminated completely from the dentin primers, the possibility of its side effects on skin tissue could not be neglected completely because it remains the main component of the dentin bonding agent, which Chigira \textit{et al.} reported as contraction gap-free\(^{21}\).

The purpose of the present study was to examine the possibility of establishing a contraction gap-free dentin bonding system which does not contain 2-HEMA.

**MATERIALS AND METHODS**

The materials employed are listed in Table 1.

The efficacy of these experimental dentin bonding systems was evaluated by measuring the wall-to-wall polymerization contraction gap width of a commercial light-activated resin composite filled into a cylindrical dentin cavity prepared in the dentin of extracted human molars.

The proximal enamel of the extracted human molar, which was stored in tap water in a refrigerator for a maximum of four weeks after extraction, was flatly

<table>
<thead>
<tr>
<th>Table 1 Dentin bonding systems tested</th>
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<tr>
<td><strong>Code</strong></td>
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<tr>
<td>Dentin conditioner 0.5 mol/1 EDTA (ph 7.4)</td>
</tr>
<tr>
<td>Dentin primer 35% 2-HEMA</td>
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<tr>
<td>35% GM</td>
</tr>
<tr>
<td>62.5% EG</td>
</tr>
<tr>
<td>45% HD</td>
</tr>
<tr>
<td>Dentin bonding agent Clearfil photo bond (CPB)</td>
</tr>
<tr>
<td>CPB without HEMA</td>
</tr>
<tr>
<td>Composite resin Silux Plus</td>
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2-HEMA; 2-hydroxyethyl methacrylate, Merck, Darmstadt, Germany
GM ; glyceryl \textit{mono}-methacrylate, Nippon Oil and Fat, Tokyo, Japan
EG ; ethylene glycol, Wako Pure Chemical Industries, Tokyo Japan
HD ; 1,6-hexanediol, Wako Pure Chemical Industries, Tokyo Japan
eliminated with carborandum paper, grit number 220. A cylindrical cavity approximately 3.0 mm in diameter and 1.5 mm in depth was prepared in the exposed dentin. The cavity wall was conditioned with 0.5 mol/L ethylenediamine tetraacetic acid (EDTA) for 60 sec which was neutralized to pH 7.4 by a NaOH solution followed by rinse and dry. The cavity was then primed with one of the experimental dentin primers for 60 sec and the cavity was dried completely. The commercial (Clearfil Photo Bond (CPB), Kuraray, Osaka, Japan) or the experimental 2-HEMA-free dentin bonding agent was applied in the cavity and irradiated for 10 sec by using a lamp unit (White Light; Takarabelmont Co., Osaka, Japan) after eliminating any excess material with a blast of air. The cavity was then filled with the commercial light-activated resin composite (Silux Plus, 3M, MN, USA) and the surface of the resin composite was momentarily flatly pressed with a glass plate and mediated with a translucent plastic matrix prior to irradiation for 40 sec. After storing the specimen in water at a room temperature of 24±1°C for 10 min, the cavity margin was exposed and the over-filled resin composite eliminated with wet carborandum paper, grit number 1000. The composite surface and surrounding dentin surface was polished with a linen cloth mediated with alumina slurry grain of size 0.3µm. The specimens were stored in water during the measuring. Marginal integrity was inspected under a light microscope. The width of any contraction gap was measured with a screw micrometer (Eyepiece Digital; Leitz, Wetzlar, Germany) mounted on a light microscope (Metaloplan; Leitz, Wetzlar, Germany) at eight points every 45 degrees along the cavity margin; the contraction gap values were presented by the sum of the diametrically opposing gap width as a percentage of the cavity diameter. The maximum of four gap values was recorded as the maximum contraction gap of the specimen. In the control group the dentin priming step was omitted and the dentin bonding agent was applied after EDTA conditioning. Ten specimens for each group, 100 in total, were prepared.

RESULTS

The contraction gap values measured are listed in Table 2.

Contraction gap formation was prevented completely with the EDTA-conditioning followed by GM, EG and HD priming despite the 2-HEMA content in the dentin bonding agent. However, a gap was observed in the 2-HEMA-priming groups which were followed by either the 2-HEMA or 2-HEMA-free dentin bonding agents. By Fisher's statistical analysis using (p<0.05), the tested groups were classified into three groups; six gap-free groups, two 2-HEMA priming groups and two no-priming groups. Therefore, it was found that the priming effect of 2-HEMA was incomplete and the 2-HEMA in the dentin bonding agent did not influence the efficacy of the dentin bonding systems tested.
2-HEMA-FREE DENTIN BONDING SYSTEM

Table 2  Contraction gap(%) of SP in the cylindrical dentin cavity

<table>
<thead>
<tr>
<th>Dentin primer</th>
<th>CPB</th>
<th>CPB without 2-HEMA</th>
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<tbody>
<tr>
<td>2-HEMA</td>
<td>0.034±0.079 (7)</td>
<td>0.032±0.045 (6)</td>
</tr>
<tr>
<td>GM</td>
<td>0 (10)</td>
<td>0 (10)</td>
</tr>
<tr>
<td>EG</td>
<td>0 (10)</td>
<td>0 (10)</td>
</tr>
<tr>
<td>HD</td>
<td>0 (10)</td>
<td>0 (10)</td>
</tr>
<tr>
<td>without priming</td>
<td>0.213±0.121 (1)</td>
<td>0.228±0.120 (0)</td>
</tr>
</tbody>
</table>

Mean±SD of the gap values. Number of gap-free specimens are in ( ).
The dentin cavity was conditioned with 0.5 mol/L EDTA for 60 sec prior to priming.
N=10

DISCUSSION

2-HEMA has been considered to be an essential component for the dentin primer, bonding agent and self-etching dentin primer. The effect of 2-HEMA was explained by its promotion of the adhesive monomer diffusion into the profound dentin or expansion the collagen fiber exposed and collapsed by acid etching during the dentin conditioning. In addition, it has been suggested that the hydrophilic and the hydrophobic groups of 2-HEMA exhibit affinities to the dentin and the resin composite, respectively, consequently improving the bond between the dentin and the resin composite. Furthermore, 2-HEMA is said to improve the wettability of the dentin bonding agent on the dentin surface. The efficacy of the 2-HEMA priming, however, was definitely inferior to that of the GM, EM, XM and two-diol solutions. As demonstrated by Chigira et al., the priming effect of GM was characterized by a high density zone on the superficial dentin layer which might indicate a monomer-concentrated layer at the adhesive interface. This finding suggested that the bonding between the resin and dentin cavity wall was established by the interaction between the calcium and the adhesive monomer at the adhesive interface. Therefore, the 2-HEMA primer had an insufficient effect because the diffusion of the functional monomer and 2-HEMA itself into the dentin was too rapid, resulting in poor monomer concentration at the adhesive interface.

However, it has been reported that this monomer has caused serious contact dermatitis as a delayed allergic reaction on the skin tissue after repeated direct contact, even when the skin was protected by rubber gloves. This side effect of 2-HEMA is considered to be caused by penetration of 2-HEMA itself into the skin tissue and absorption in the T-cells, resulting in the allergic reaction. To avoid such a side effect, we would recommend the use of esterified methacrylate with polyvalent alcohol and diol solutions as the contraction gap-free dentin primer, although the detailed mechanism of these primers have not been completely clarified.

As demonstrated in this study, the combination of the dentin conditioner of EDTA, three primers composed of aqueous solution of GM, EG or HD and the commercial dentin bonding agent containing 10-methacryloxydecyl dihydrogen phos-
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Phosphate (10-MDP) completely prevents contraction gap formation in the light-activated resin composite in a cylindrical dentin cavity. Furthermore, it was noteworthy that gap formation was prevented completely regardless of the 2-HEMA content of the dentin bonding agent.

The dual-cured dentin bonding agent tested in this study was prepared by mixing two drops, one containing 10-MDP, bis phenol A diglycidyl methacrylate (Bis-GMA), 2-HEMA and the other containing sodium sulfate, ethanol and polymerization activators. The ethanol in this bonding agent possibly improved the penetration and the wettability of the adhesive monomer to the dentin cavity wall, providing the same function as 2-HEMA.

The side effects on skin tissue of 2-HEMA could be avoided by using 2-HEMA-free dentin bonding system introduced in this study. However, it was revealed that primary irritation of skin tissue was serious with 10-MDP whereas that of 2-HEMA was negligible. This side effect of 10-MDP was reported by observation of the oral mucous which exhibited a slight inflammatory reaction after the dentin bonding agent application. More effort is required to develop a dentin bonding system with completely negligible side effects.

REFERENCES


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