Effect of 2-hydroxyethyl methacrylate in an Al_2Ox_3/glycine solution used as primer for dentin bonding

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The aim of this study was to investigate the effect of 2-hydroxyethyl methacrylate (HEMA) in an Al_2Ox_3/glycine solution used as primer for dentin bonding in combination with an experimental BISGMA–HEMA-based adhesive resin. With various amounts of HEMA in the Al_2Ox_3/glycine solution, a shear bond test showed a maximum strength at 50 wt% of HEMA. In this case, scanning electron microscopy examination showed a HEMA-infiltrated surface layer of the dentin along the interface between dentin and the adhesive resin. It is suggested that HEMA in Al_2Ox_3/glycine solutions may facilitate penetration of HEMA into etched or primed dentin. □ *Adhesive resin; dentin substrate; scanning electron microscopy; shear bond*

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It has been observed that 2-hydroxyethyl methacrylate (HEMA) may mediate a bonding reaction between dentin and composite resin. Gluma, an aqueous solution of 35% HEMA and glutaraldehyde, has been found to enhance the bond between composite resin and dentin (1). It has also been demonstrated that this agent gives rise to a good bond to dentin in combination with various commercial dentin-bonding agents (2). Itoh & Wakumoto (3) reported that the pretreatment of dentin walls with 35% HEMA solution improved the marginal adaptation.

Recently, utilizing the compatibility of HEMA and dentin, several attempts have been made to combine the steps of dentin pretreatment to simplify the clinical procedure for dentin bonding. Some investigators have reported that a self-etching dentin primer, which contained an acid or an acidic monomer in 35% HEMA solution, mediated an acceptable bond between dentin and composite resin (4, 5). De Araujo & Asmussen (6, 7) have presented a simplified Gluma system, in which an aluminum oxalate solution containing glycine (Al_2Ox_3/Gly; pH 1.25) and Gluma resin are used as the dentin primer and dentin-bonding agent, respectively. A mixture of Al_2Ox_3/Gly and HEMA may therefore perform as a self-etching dentin primer.

The purpose of this study was to investigate the effect of HEMA in an Al_2Ox_3/Gly primer on the strength of the bond to dentin obtained with an experimental bonding agent containing BISGMA, HEMA, and propionic aldehyde. The composition of the adhesive resin was chosen so as to approximate the composition of Scotchbond 2 with regard to BISGMA and HEMA, and that of Gluma with regard to aldehyde. The primed dentin surface was also examined under scanning electron microscope (SEM).

Materials and methods

The materials used in the study are listed in Table 1. Al_2Ox_2/Gly solutions (pH 1.25) containing 0, 10, 20, 30, 40, 50, 60, or 70 weight % of HEMA was used as primer of the dentin surface. The composition of the experimental adhesive resin is also shown in Table 1.

Table 1. Composition of primer and adhesive resin used in the study

Primer	
Al_2Ox_3/Gly solution containing HEMA	
Aluminum nitrate	4.96 g
Oxalic acid	2.50 g
in 100 g H_2O	
pH adjusted to 1.25 with glycine	
HEMA added to 0, 10, 20, 30, 40, 50, 60, or 70 wt%	
Adhesive resin	
BISGMA	25 mol%
HEMA	74 mol%
Propionic aldehyde	1 mol%
with 0.2 wt% camphorquinone	
and 0.2 wt% *N,N*-cyanoethylmethylaniline (CEMA)	

Shear bond strength

The investigation was carried out on human molar teeth extracted from the permanent dentition. After mechanical cleaning, the tooth was embedded in epoxy resin. A flat surface of dentin was produced by wet grinding on carborundum paper no. 1000 at the final stage. The conditions of the treatment of dentin are summarized in Table 2, presenting the results. As standard procedure, the dentin surface was primed with Al_2Ox_3/Gly solution containing various amounts of HEMA for 60 sec. After being dried with a stream of air (rinse (−)), the adhesive resin (Table 1) was applied with a small disposable brush and then again exposed to a weak stream of air until a thin resinous residue remained on the dentin surface. Thereafter, this resinous layer was preirradiated for 20 sec with a light source (Luxor, ICI, U.K.). Two more groups were prepared by the use of 50% HEMA in Al_2Ox_3/Gly: in the first group rinsing was done with distilled water for 10 sec before the adhesive resin was applied (rinse (+)). In the other group, preirradiation of the adhesive resin was omitted (light (−)). A split Teflon mold with a cylindrical hole (height, 2.5 mm; diameter, 3.5 mm) was clamped on the treated dentin surface as previously described (8). Silux plus (3M, USA) was filled into the hole by means of a syringe and irradiated with the light source for 60 sec. After 24 h of storage in water at 37 °C, the specimen was mounted in a mechanical testing machine (Instron Ltd., High Wycombe, U.K.), and the shear bond strength was measured at a crosshead speed of 1 mm/min. Six specimens were prepared for each condition of treatment.

Table 2. Shear bond strength to dentin

Primer		Preirradiation	Shear bond strength
HEMA	Rinse	(20 sec)	(MPa)
0%	−	+	3.8 ± 1.1
10%	−	+	6.2 ± 1.9
20%	−	+	9.7 ± 7.1
30%	−	+	12.3 ± 3.4
40%	−	+	13.4 ± 4.0
50%	−	+	17.3 ± 4.8
	+	+	7.1 ± 1.3
	−	−	5.8 ± 3.9
60%	−	+	16.4 ± 3.8
70%	−	+	8.4 ± 3.5

SEM examination

Several flat dentin surfaces primed with 0 or 50% HEMA in Al_2Ox_3/Gly (rinse (−)) were prepared after air-drying as mentioned above. Specimens of the bonding interface between resin and dentin were prepared as follows. The shear bond specimens obtained by the standard procedure by use of 50% HEMA in Al_2Ox_3/Gly were longitudinally sectioned through the center of the composite resin part with a slow-speed diamond

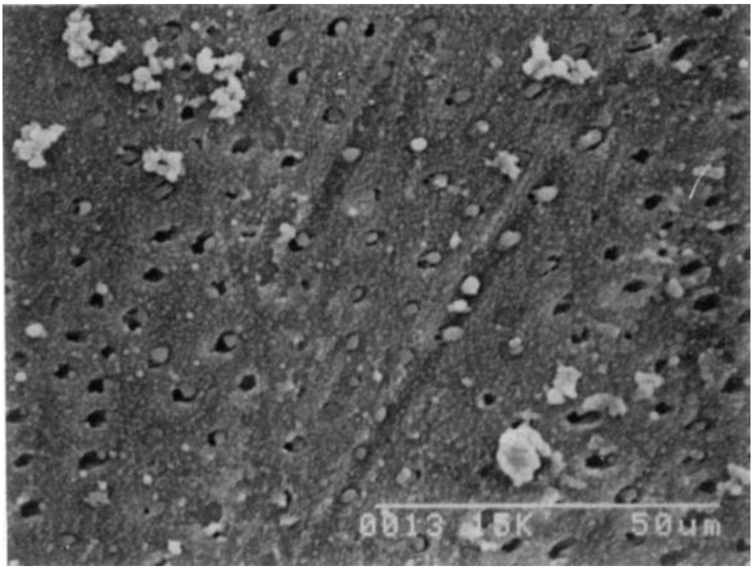

Fig. 1. Dentin surface primed with 0% HEMA in Al_2Ox_3/Gly solution (rinse (−)). Several small particles are found on the surface. Obturation of dentinal tubules can be seen.

saw under water. After being rinsed with distilled water and air-dried, the sectioned surfaces were etched with 6 *N* HCl for 10 sec, rinsed, and dried. Two specimens were examined in each group.

Statistical analyses

The relationship between HEMA concentration and shear bond strength was subjected to regression analysis. The additional groups with 50% HEMA were compared by means of Student's *t* test.

Results

Shear bond strength

The results are shown in Table 2. Without rinsing and after priming with the various Al_2Ox_3/Gly solutions, the strength increased from 3.8 to 17.3 MPa when the concentration of HEMA rose from 0 to 50%. In this range a significant correlation was found between the concentration of HEMA and shear bond strength ($r = 0.727$; $p < 0.001$). With the concentration of HEMA above 50% the strength decreased significantly to 8.4 MPa at 70% HEMA ($r = -0.637$; $p < 0.005$). The additional groups with 50% HEMA in Al_2Ox_3/Gly, rinse (+) or light (−), showed significantly lower strength than the standard group ($p < 0.005$).

SEM examination

Examples are shown in Figs. 1–3. After priming with 0% HEMA (Fig. 1) several small particles were found on the dentin surface. Obturation of dentinal tubules was observed. The dentin surface primed with 50% HEMA appeared smoother than that treated with 0%, and only a few particles were seen (Fig. 2). A significant number of dentinal tubules were obturated also in this case. At the bonding interface a thin acid-resistant layer 2–3 μm thick was observed between dentin and adhesive resin (Fig. 3). This layer seemed to penetrate the dentin.

Discussion

The present study examined various concentrations of HEMA in an Al_2Ox_3/Gly solution used as primer in dentin bonding. The results show that the effect of HEMA on bonding to dentin depends on its concentration in the Al_2Ox_3/Gly solution. At

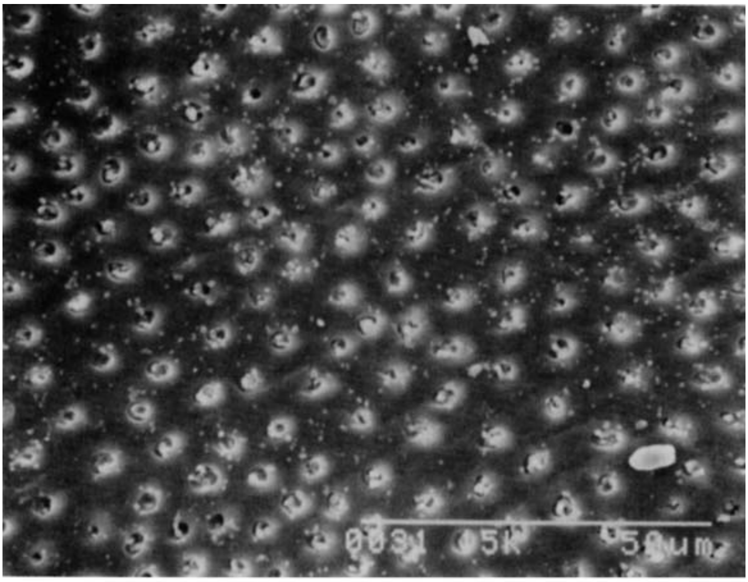

Fig. 2. Dentin surface primed with 50% HEMA in Al_2Ox_3/Gly solution (rinse (−)). It appears smoother than in Fig. 1. Most dentinal tubules are obturated.

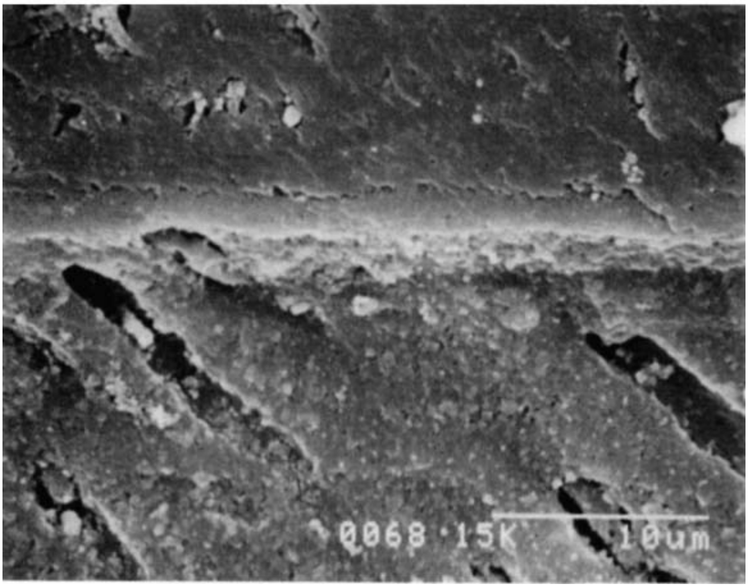

Fig. 3. The bonding interface between adhesive resin and dentin primed with 50% HEMA in Al_2Ox_3/Gly solution (rinse (−)). A thin acid-resistant layer 2–3 μm thick is observed between adhesive resin (top) and dentin (bottom).

about 50% HEMA the shear bond strength reached a maximum value. At this concentration the examination by SEM showed a smoother dentin surface than that obtained with 0% HEMA in the primer. This observation may be related to the adsorption of HEMA into dentin. Harashima & Hirasawa (9) have found that HEMA can infiltrate into intertubular dentin. The thin acid-resistant layer observed at the bonding interface may indicate the infiltration of HEMA. This HEMA-infiltrated layer mediates a good bond between dentin and the adhesive resin. Therefore, the nature of this layer would affect the effectiveness of the bonding procedure. The low strength of the bond in the two additional groups with 50% HEMA may be explained by the poor quality of the layer. In his study with Scotchbond 2 and Gluma Erickson (10) found that an effective bonding to dentin is dependent on infiltration and polymerization of resin in the dentinal matrix. Rinsing the primed surface must have removed the adsorbed HEMA. Besides, preirradiation may have resulted in a greater extent of conversion within the HEMA-infiltrated layer and the adhesive resin, leading to greater strength of the bond.

When the HEMA concentration was more than 50%, the shear bond strength decreased. There may be several explanations of this phenomenon. The first is a saturation of the dentin surface by HEMA. Harashima & Hirasawa (9) have reported that the adsorption of HEMA in dentin has a maximum point. Second, as the concentration of HEMA is increased, the concentration of Al_2Ox_3/Gly is decreased. At decreased concentrations of Al_2Ox_3/Gly the primer may not be able to modify the dentin so thoroughly as to supply enough space for the subsequent penetration of HEMA. Furthermore, the variation in bond strength with HEMA may be associated with the affinity between the modified dentin and the adhesive resin. The affinity between a polymeric surface and a liquid involves not only their surface energy but also the solubility parameter and polarity (11). Further investigations of the affinity between primed dentin and adhesive resin will be necessary to assess the bonding mechanism.

The observed obturation of dentinal tubules is in agreement with the findings of De Araujo & Asmussen (6) and may be associated with a precipitation of insoluble Ca-oxalate and Al-phosphate (12).

To summarize, primers containing HEMA in an Al_2Ox_3/glycine solution may modify the surface of dentin in such a manner that the result is an increased strength of the bond to restorative resins. The influence of HEMA on the efficacy of adhesive resins other than the one used in the present study is not known. Possibly, the adhesive resin should also contain HEMA to have an affinity suitable for bonding to the modified surface.

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