Penetration of restorative resins into acid etched enamel. II

Dissolution of entrapped air in restorative resin monomers

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Viscosity, surface tension and contact angle are factors that influence the penetration of restorative resins into acid etched enamel. Furthermore, as the resin is drawn by the capillary forces into the pores of the etched enamel the pressure of the entrapped air will increase. The increased pressure has the effect that air will dissolve in a resin that is saturated with air at one atmosphere. The purpose of the present work was 1) to investigate the rate of dissolution of included air bubbles at increased pressure, 2) to use the results to calculate the depth of penetration by means of a cylindrical model of the capillary pores, and 3) to check the results of the calculations by measurements of the tag lengths of restorative resins placed on acid etched enamel. The rate of dissolution was measured in monomers of varying viscosity in a glass syringe by means of a stereo microscope. The calculations showed that the depth of penetration decreases only slightly with viscosity. Thin sections of restorative resins placed on acid etched enamel were prepared whereafter the enamel was dissolved in hydrochloric acid. Tag lengths of 50 μm or more were observed with composite as well as non-composite resins.

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Viscosity, surface tension and contact angle are factors that influence the penetration of restorative resins into acid etched enamel. These factors were investigated by *Asmussen* (1977) for a number of restorative resin monomers. When a restorative resin is applied to an etched enamel surface, air will be trapped in the pores of the surface. The depth of penetration of the resin may hereby be reduced. It is, however, possible that the entrapped air will dissolve in the resin monomer. Initially the monomer is saturated with air at a pressure of one atmosphere. As the resin is drawn by the capillary forces into the pores, the pressure of the entrapped air will increase. According to Henry's law the

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solubility of air in the monomer will hereby also increase. Consequently it seems possible that the depth of penetration could be determined by the rate of dissolution of air in the monomer. It was the purpose of the present work 1) to investigate the rate of dissolution of included air bubbles at increased pressure in restorative resin monomers, 2) to use the results together with the results obtained by *Asmussen* (1977) to calculate the depth of penetration by means of a cylindrical model of the capillary pores, and finally 3) to check the results of the calculations by measurements of tag lengths of restorative resins in acid etched human enamel.

MATERIALS AND METHODS

The experimental part of the present study was divided into two sections in which the dissolution of air bubbles and the tag lengths were investigated.

Rate of dissolution of air bubbles in resin monomers at increased pressure

The measurements were carried out in three different monomer mixtures. The composition and the viscosity at 23 °C of the mixtures are presented in Table I. Similar monomer mixtures are encountered in commercial brands of restorative resins (*Asmussen*, 1975). The viscosities in Table I were obtained from the paper of *Asmussen* (1977). Air bubbles in a monomer mixture were produced by stirring with a spatula for 30–45 seconds. Immediately thereafter about 0.5 ml of the monomer mixture was sucked into a 1 ml glass syringe with a 4.61 mm internal diameter, the tip of which was then stoppered. The piston was removed, and approximately 0.2 ml of water was placed on top of the mixture, whereafter the piston was replaced in the syringe. The water was introduced 1) to avoid polymerization of the monomer in the narrow space between piston and the walls of the syringe and 2) to obtain the same friction between piston and the walls when using the three monomer mixtures. With water in the unstoppered syringe this friction was approx. 75 g. The syringe was hereafter placed in a horizontal position on a stand in a stereo microscope fitted with a calibrated ocular. The nominal magnification was 8x18. The piston was loaded with 250, 500, 750 or 1000 g force, respectively. After subtracting the friction from these loads the applied pressures were calculated to be 1.03×10^6, 2.50×10^6, 3.97×10^6 or 5.44×10^6 dyne/cm^2. After a pressure had been applied to a mixture, an air bubble was quickly located in the microscope, and the diameter of the bubble was recorded in whole ocular scale divisions (1 div. = 6.25 μm). The time elapsed from this measurement until the bubble had disappeared, was determined by means of a stop watch. After the bubble had dissolved completely, the load was removed from the piston for 1 minute. The measuring procedure was repeated on new air bubbles until 25 min after the mixture had been stirred. Then the syringe was emptied, cleaned, and the measurements continued in a new mixture. The diameter of the recorded bubbles was in the range from 13 to 63 μm. For each monomer mixture and pressure the time of disappearance was measured for five bubbles of a given diameter.

An analysis of the results showed that the time of disappearance increased statistically

Table I. *Composition and viscosity of monomer mixtures used in section A of the investigation*

Monomer mixture	BIS-GMA (mole %)	TEDMA (mole %)	Viscosity (cP) at 23 °C
A	80	20	40500
B	60	40	3200
C	40	60	380

BIS-GMA: Bisphenol-A-glycidyl-dimethacrylate
TEDMA: Triethylene-glycol-dimethacrylate

significantly with the time of the measurement. However, the contribution of this increase to the total variation was small, and it was therefore disregarded in the following.

Tag lengths of restorative resins placed on acid etched enamel

The materials listed in Table II were investigated. Cariesfree enamel surfaces of recently extracted human teeth that had been stored in 1% chloramine until use were cleaned with an aqueous slurry of pumice by means of a tooth brush. The teeth were rinsed in demineralized water, dried by an air blast, and etched for 1 minute with 35% H_3PO_4. The acid was applied with a cotton pellet. The etched surfaces were now rinsed for 15 s in demineralized water and dried anew for 10 s. The restorative resins were applied immediately after mixing to the etched enamel surface and covered with a lightly tightened matrix band. From each material five specimens were prepared. After setting of the restorative resin the coated teeth were embedded in an epoxy resin and sectioned longitudinally, approx. perpendicularly to the enamel surface, by means of a Bronwill Thin Sectioning Machine, Universal Model 77 (Bronwill Scientific, Inc., Rochester, N.Y., USA). The thickness of the sections was about 110 µm. Two sections of each tooth were fixed between a microscope objective and cover glass that then were glued together with Canada balsam in two peripheral areas apart from the sections. These assemblies were immersed in 5% hydrochloric acid until the enamel had dissolved completely. The tags were observed and measured in transmitted light in a microscope fitted with a measuring ocular. The nominal magnification of the microscope was 80 x 0.8 x 8. The investigation was carried out at a room temperature of 23 ± 2 °C. The possibility that the observed tags were residues from the enamel was assessed by immersing a number of specimens in a basic solution of hypochlorite until the remnants of the dentin had dissolved completely. It was found that the tags were not effected by this treatment.

Table II. *Materials used in section B of the investigation*

Brand	Batch No.	Manufacturer
Concise Enamel Bond	6020 B1	3 M Company, Minnesota, USA
Concise	6086 F31	3 M Company, Minnesota, USA
Adaptic	3 G 027	Johnson & Johnson, New Jersey, USA

RESULTS

Rate of dissolution of air bubbles

For each monomer mixture and pressure the logarithm of the time of disappearance was plotted against the logarithm of the diameter of the bubbles. The points were distributed about a straight line, and a regression analysis was performed to determine the parameters of the line. The straight line relationship between the logarithms of the variables was then transformed to obtain the relationship between the variables. This relationship is of the form $T = a \cdot D^b$, where T is the time of disappearance in seconds, and D the diameter of the bubbles in µm. The calculated «constants» a and b are given in Table III for the respective monomer mixtures and pressures.

Tag lengths

Tag lengths of 50 µm or more were observed for all three brands. No difference in tag lengths or frequency of tags could be observed between the brands, but the lengths of tags were found to vary considerably from tooth to

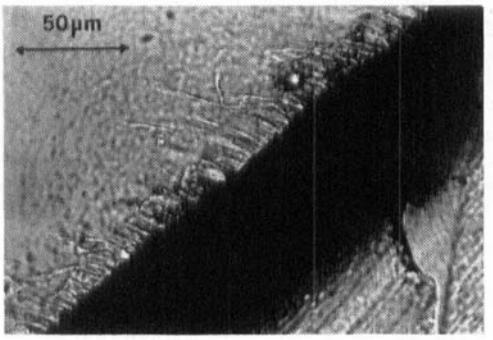

Fig. 1. Tags from Concise Enamel Bond.

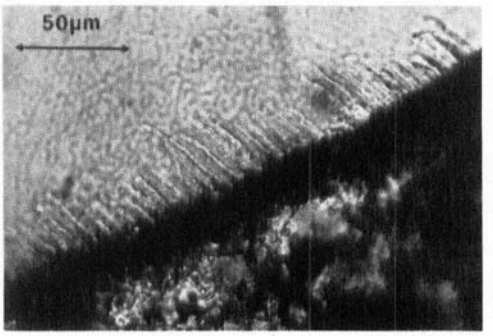

Fig. 2. Tags from Concise.

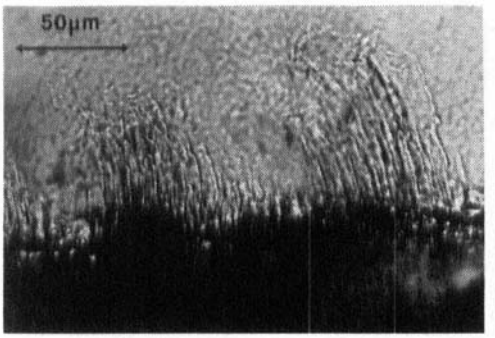

Fig. 3. Tags from Adaptic.

tooth. Figs. 1, 2 and 3 show tags formed by Concise Enamel Bond, Concise and Adaptic, respectively.

DISCUSSION

The results obtained on the dissolution of air in the present study and those by *Asmussen* (1977) may be used to calculate the depth to which a given monomer will penetrate into the pores of an etched enamel surface. If a cylindrical model of the pores in the etched enamel is assumed, the process of penetration may be analyzed on the basis of Fig. 4. The resin R has penetrated to the depth l into the capillary cylinder C, of length l_0 and diameter d. The contact angle is θ. Due to the curvature of the resin-air interface a pressure difference

$$P_2 - P_1 = \frac{4\gamma \cdot \cos\theta}{d}$$

occurs between the entrapped air at pressure P_2 and the resin just below the surface at pressure P_1. γ is the surface tension of the liquid. As the resin is drawn by the capillary forces into the pore, the pressure P_2 of the entrapped air increases according to Boyle-Mariotte's law and is given by $P_2(l_0 - l) = P_0 \times l_0$. P_0 is the normal atmospheric pressure. The pressure difference ΔP that causes the capillary penetration is $P_0 - P_1$, which according to the two above equations may be expressed as

$$\Delta P = \frac{4\gamma \cos\theta}{d} - P_0 \cdot \frac{l}{l_0 - l}$$

From Poiseuille's equation the relation between the differentials dt and dl is obtained as

$$dt = \frac{32\eta \cdot l}{d^2 \cdot \Delta P} \cdot dl$$

where t is the time and η is the viscosity of the liquid. From this equation the time necessary to penetrate to a given depth may be calculated by numerical integration.

Next, assume that the resin has penetrated according to equation (1) to the maximum depth l_1, determined from $\Delta P = 0$. At this depth of penetration the overpressure of the entrapped air is

$$\frac{4\gamma \cos\theta}{d}$$

It may be demonstrated that the time necessary to penetrate the distance Δl by dissolution of air at a given overpressure is approximated by the time of disappearance at the same overpressure of an air bubble of diameter $2 \cdot \Delta l$. Thus, the penetration Δl by dissolution in a given time T may be determined from an equation of the type $T = a \cdot (2 \cdot \Delta l)^b$, in which the constants a and b depend upon the overpressure and the viscosity of the monomer. The constants a and b at given overpressures and monomer viscosities may be obtained by means of Table III if it is assumed that a linear relationship exists between log T (D constant) and overpressure, as well as between log D (T constant) and log η. These assumptions of approximate linearity were justified from a number of plots obtained by means of the constants listed in Table III and the viscosities listed in Table I.

On the basis of the above considerations the depth of penetration during a given time can be calculated. A pore length of 50 μm was assumed, and the calculations were carried out for pore diameters of 1 μm and 0.4 μm, respectively. The time necessary to penetrate to the depth l_1 solely by capillary attraction is of course infinite; but the time t_1 necessary to penetrate to the depth l_1 - 1 μm can be obtained from equation (1) by numerical integration.

Table III. *Calculated constants a and b in the relation $T = a \cdot D^b$ between time of disappearance T (sec.), and diameter of air bubles D (μm)*

Monomer mixture		Pressure (10^6 dyne/cm^2)			
		1.03	2.50	3.97	5.44
A	a	0.707	0.911	0.601	0.343
	b	1.951	1.665	1.709	1.809
B	a	0.0446	0.0589	0.102	0.0299
	b	2.472	2.158	1.928	2.213
C	a	0.00674	0.00289	0.00125	0.00737
	b	2.537	2.626	2.767	2.255

This was done for monomers of the following viscosities: 100, 380, 1000, 3200, 10 000 and 40 500 cP. The corresponding values of γ and cos θ were obtained from the paper of *Asmussen* (1977). The period of time in which the monomer is able to flow being t_0, the additional depth of penetration by dissolution of the entrapped air during the time $t_0 - t_1$ was calculated in the manner described above. In the calculations the overpressure of the entrapped air was assumed to be

$$\frac{4\gamma \cos\theta}{d}$$

and t_0- values of 30, 60 and 120 seconds were used.

The results of the calculations are presented in Figs. 5 and 6, in which the total depth of penetration during 30, 60 or 120 seconds is plotted against the logarithm of the viscosity. In Fig. 5 the pore diameter is 1 μm, and in Fig. 6 the diameter is 0.4 μm. The upper horizontal line indicates the pore length of 50 μm, the lower horizontal line the penetration depth l_1. It appears from the figures 1) that for given penetration times, a complete penetration is obtained for monomers with viscosities below a certain value, and 2) that the depth of penetration decreases only slightly with viscosities above this critical value. The calculations are supported by the results of the tag measurements. The viscosities at

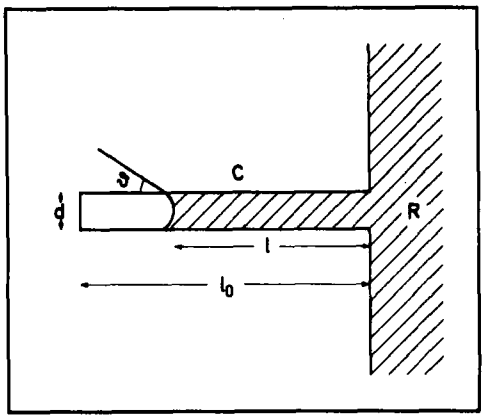

Fig. 4. Schematic presentation of the penetration of resin into the pores of an etched enamel surface. The pores are represented by the capillary cylinder C, of length lo and diameter d. The resin R has penetrated to the depth 1, θ is the contact angle.

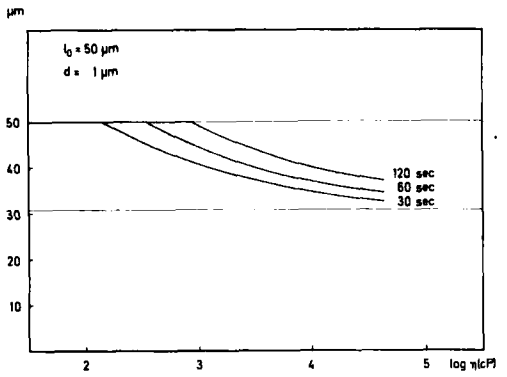

Fig. 5. Calculated depth of penetration (µm) during 30, 60 or 120 seconds in relation to the logarithm of resin viscosity (cP). The length and diameter of the cylindrical pore was 50 µm and 1 µm, respectively. The upper horizontal line represents the pore length, the lower horizontal line the maximum depth of penetration attainable solely by capillary attraction.

23 °C of the monomers in the investigated brands are 280, 5100 and 13 700 cP, respectively (*Asmussen*, 1977), and in all cases tag lengths of 50 µm or more were observed.

This finding is in agreement with the findings of *Jørgensen & Shimokobe* (1975) and *Pahlavan, Dennison & Charbeneau* (1976). With acid etched enamel these authors found no significant differences between low-viscous, non-composite resins and composite resins, in terms of adaptation and depth of penetration.

However, the above results contrast with the results obtained by *Dogon* (1976). *Dogon* found that the depth of penetration into acid etched enamel was considerably reduced when the viscosity of the resin was above 500 cP. In this investigation the viscosity of the resin was changed by adding varying amounts of silanized Cab-O-Sil of a particle size of 0.01 µm to Concise Enamel Bond. Suspensions of this type are known to be thixotropic when the concentration of powder is raised above a certain level. This means, among other things, that a shear stress limit exists, below which the suspension will not flow. Thus, it could be conceived that this is the explanation why the depth of penetration was reduced when 6% by weight or more of Cab-O-Sil was added to the Concise Enamel Bond. To investigate this possibility the present author produced mixtures of varying viscosities by adding 0, 10, 20, 30% by weight of silanized Aerosil of a particle size of 0.016 µm to the low-viscous, non-composite fissure sealant Delton® Johnson & Johnson). The depth of penetration was measured with these mixtures using the method described above. In all cases tag lengths of 50 µm were observed. Thus, the results obtained by *Dogon* could be neither confirmed nor explained by the present author.

To summarize, on the basis of the calculations and experimental results of the present work it may be concluded that an intermediate layer of low-viscous, non-composite resin between composite restorative and etched enamel is not likely to improve the quality of composite restorations from what may be obtained by applying the composite material directly to the etched enamel surface.

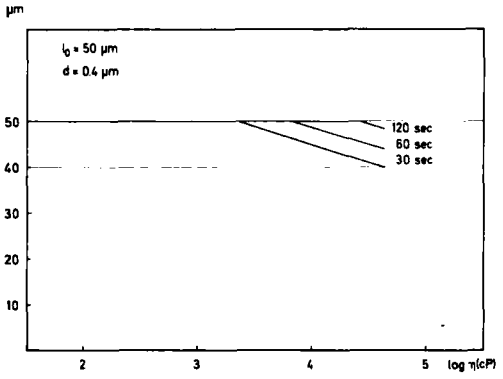

Fig. 6. Calculated depth of penetration (µm) during 30, 60 or 120 seconds in relation to the logarithm of resin viscosity (cP). The length and diameter of the cylindrical pore was 50 µm and 0.4 µm, respectively. The upper horizontal line represents the pore length, the lower horizontal line the maximum depth of penetration attainable solely by capillary attraction.

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