Adhesive bonding of dental luting cements; influence of surface treatment

GUDBRAND ØILO

NIOM – Scandinavian Institute of Dental Materials, Oslo, Norway and Department of Prosthetic Dentistry, Dental Faculty, University of Oslo, Oslo, Norway

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Tensile bond strength of four different luting cements to smooth dentin surfaces was measured. A chisel edged, stainless steel ring was cemented to the butt end of a dentin cylinder. The dentin was polished to a plane and smooth surface before cementation. The cements were also applied to dentin surfaces that were treated with a pumice slurry, etched with different acid solutions, or covered with different liners.

The results showed that the polycarboxylate cement had a tensile bond strength to smooth, untreated dentin of approximately 4 MN/m^2. The zinc phosphate and EBA cements had a bond strength of 0,6 MN/m^2 and the composite resin cement had no measurable bond to untreated dentin.

All dentin treatments showed in general a decreasing effect on the bond strength of zinc phosphate, polycarboxylate and EBA cements, whereas that of composite resin cement showed a slight increase.

Key-words: Dental materials; dentin; acid etch; scanning electron microscopy

Gudbrand Øilo, Department of Prosthetic Dentistry, Dental Faculty, University of Oslo, Geitmyrsveien 69, Oslo 4, Norway

The tensile bond strength of dental restorative materials to hard dental tissues has attained considerable interest as an important factor for the sealing effect and the retention of restorations. The bond to enamel has been studied extensively and a good mechanical retention has been obtained with several resin based materials (4, 6). The bond to dentin, however, still remains a problem with the majority of materials used (4).

The bond strength of dental luting cements has been studied in several investigations, and it has been shown that the polycarboxylate cement is the only cement with significant adhesion to dentin (17, 15). Several surface treatments of the dentin have been tried in order to increase the bond strength of cements, and it has been shown that acid etching could increase the bond for several resin cements to bovine dentin (8, 13), a layer of varnish, however, seemed to reduce the bond for all types of cements (7).

It was the purpose of the present investigation to compare the tensile bond strength of different cements under standardized conditions, to study the influence of several surface treatments on the surface structure of the dentin and the influence of these treatments on the bond strength of the cements.

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MATERIALS AND METHOD

The cements listed in Table 1 were mixed at 23 ± 1 °C and according to the manufacturers instructions.

Recently extracted erupted and unerupted molars were embedded in resin (Epofix, Struers Scientific Instruments, Copenhagen, Denmark). The enamel was cut off the occlusal surface, and the remaining crown was turned down to a dentin cylinder 5 mm in diameter (Fig. 1). The tooth specimen was then mounted in a holder and the butt end of the dentinal cylinder was polished at right angle to the mid-axis. Polishing took place on wet carborundum paper no. 800 (Struers Scientific Instruments, Copenhagen, Denmark). After polishing, the dentin was given one of the following treatments:

1. Sprayed with water.
2. Polished lightly on a linen cloth with pumice slurry.
3. Etched for 10 seconds with a 1 % aqueous solution of phosphoric acid.
4. Etched for 60 seconds with a 30 % aqueous solution of phosphoric acid.
5. Rinsed for 60 seconds with a cavity cleanser consisting of 45 % citric acid (10). (Lee Pharmaceuticals, South El Monte, California, U.S.A.).
6. Rinsed for 60 seconds with a cavity cleanser (Tubulicid, red label, Dental Therapeutics A.B., Nacka, Sweden) and subsequently covered with one layer of a liner (Tubulitec, Dental Therapeutics A.B., Nacka, Sweden).
7. Covered with one layer of a varnish (Copalite, Cooley & Cooley Ltd, Houston, Texas, U.S.A.).
8. Covered with one layer of an alfa-cyanoacrylate adhesive (Miracle Bond, type S, Koatsu Gas Industrial Co., Ltd., Osaka, Japan).

The tooth specimens were kept wet *during the whole* procedure, but prior to the surface treatment, the dentin was dried with air at room temperature. After the surface treatment they were kept in 100 % relative humidity until cementation. The same tooth specimen was used for several tensile bond tests, but was always ground and polished well before new surface treatment and cementation.

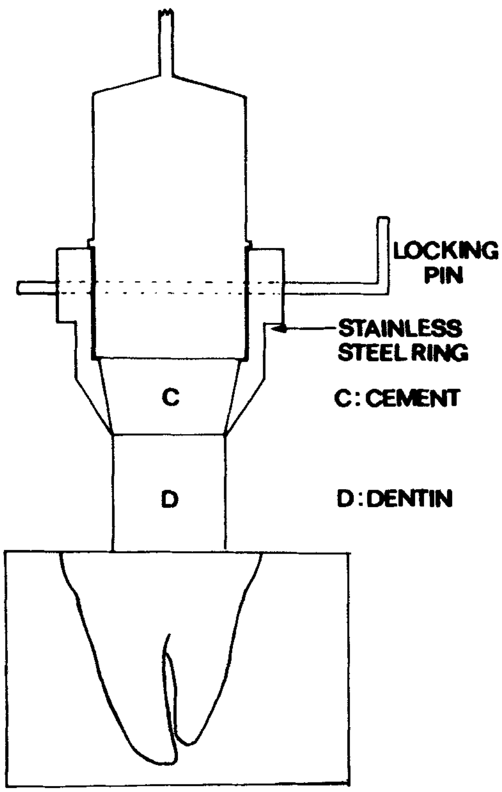

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Fig. 1. Diagram of experimental assembly.

Several samples from each surface treatment were coated with a layer of gold and studied in a scanning electron microscope (Jeol JSM-A Scanning Microscope, JEOL Ltd., Tokyo, Japan). The surface roughness was also measured on several dentin surfaces using an electronic surface profile measuring system (Perth-O-Meter, Type W3 Bc, Perthen, Hannover, Germany).

A stainless steel ring with an inner diameter of 5 mm was used as the holder for the cement. A plug was locked to one end of the ring (Fig. 1) which enabeled the con-

Table 1. *Cements used in the present study*

Type	Name	Manufacturer	Batch no.	Powder/liquid-ratio
Zinc phosphate	De Treys Zinc Cement Improved	De Trey Freres S.A. Zürich, Switzerland	Powder TD2TMK Liquid TD2TMK	3 g/ml
Polycarboxylate	Durelon	ESPE GmbH Seefeld/Overbay, Germ.	Powder P762381 Liquid LD09018	1,5 g/g
Reinforced zinc oxide eugenol	Opotow Alumina EBA, Crown & Bridge cement	Teledyne Dental Getz-Opotow Div. Elk Grove Village, Illinois, U.S.A.	112175	7 g/ml
Composite Resin	Epoxylite CBA 9080	Lee Pharmaceuticals, South El monte, California, U.S.A.	1074BP–2	2 g/g

nection to a universal testing machine (Testatron S 718, Otto Wolpert Werke GmbH, Ludwigshafen/Rhein, Germany). The other end of the ring was chisel edged with the bevel on the outside of the ring (Fig. 1). The ring with the connector plug was placed in a holder (Fig. 2) and fixed in correct position on top of the dentin cylinder. The ring was filled to excess with cement and by means of the same holder, placed in the correct position on the dentin surface. The holder with the specimen was then placed in a humidifier for 10 min. Excess cement was thereafter removed carefully while the tooth specimen and the ring remained fixed in the holder. The specimen was thereafter removed and stored in water for 24 hours at room temperature. The specimen was then mounted in the testing machine, and a tensile load was applied with a crosshead speed of 1 mm/min until rupture of the bond between cement and dentin.

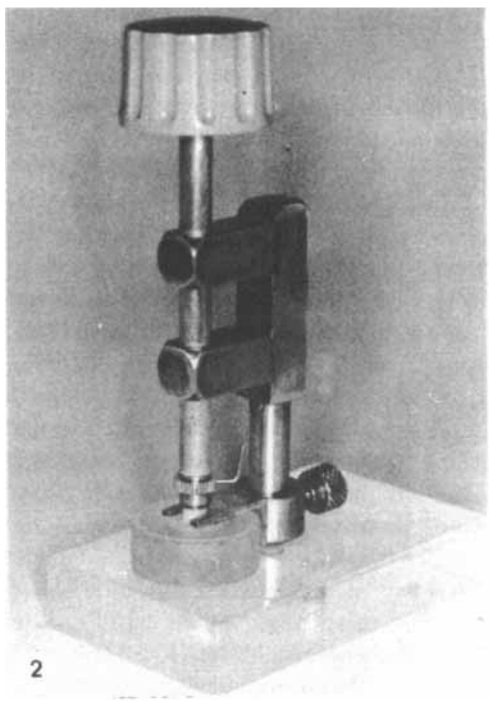

Fig. 2. Dentin cylinder and cement holder in fixation apparatus.

RESULTS

Bond strength

The tensile bond strengths of the four different cements are presented in Fig. 3. Large differences were found in the bond strength of the various cements and in the influence of the different surface treatments.

The polycarboxylate cement had a bond strength of 3.8 MN/m^2 to untreated dentin.

The break of bond was mainly a cohesive break in the cement as 70–80 % of the dentin surface was covered by a layer of remaining cement (Fig. 4).

The zinc phosphate and EBA cements had a small bond strength to smooth, untreated dentin (0.6 MN/m^2). The break of bond was interfacial. A bond between untreated dentin and composite resin cement could not be recorded.

The bond strength of zinc phosphate cement to dentin surfaces which had been cleaned with pumice or etched with an acid, was significantly lower ($P < 0.01$) than that to the untreated surfaces. The bond strength of the EBA cement was not significantly changed. The polycarboxylate cement, however, had a significantly lower bond strength to the surfaces that were etched with 30 % phosphoric acid ($P < 0.05$) or cleaned with a citric acid solution ($P < 0.01$) than to untreated dentin. An inspection of the dentin surface after break of the bond showed that very small amounts of cement adhered to the dentin (Fig. 5).

The composite resin cement showed a minor increase of the bond strength after etching with various solutions of phosphoric acid. However, it obtained values close to that of the polycarboxylate cement after a surface treatment with the citric acid solution (Fig. 3).

The application of varnish or liners to the dentin surfaces had a significant decreasing effect on the bond strength of zinc phosphate cement ($P < 0.05$). However, the adhesion to surfaces treated with the Tubulicid/Tubulitec system was significantly better than that to surfaces covered with Copalite or cyanoacrylate adhesive ($P < 0.05$). The polycarboxylate cement had no measurable bond strength to surfaces covered with the cyanoacrylate adhesive whereas the composite resin cement exhibited a small adhesion (Fig. 3).

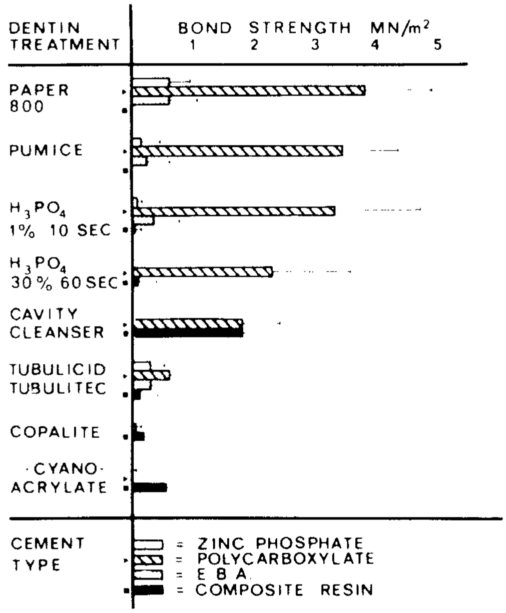

Fig. 3. Bond strength of some dental luting cements to dentin.

Surface structure

The scanning electron microscopy (SEM) of the different surfaces showed that a layer of grinding debris, which was observed on the untreated dentin (Fig. 6), was reduced by the treatment with pumice (Fig. 7) and totally removed when etching with the various acid solutions (Fig. 8, 10). A treatment with 1 % phosphoric acid in 10 seconds seemed to leave the tubules open (Fig. 8). This assumption was confirmed when studying the cement side of the interface between composite resin and etched dentin which showed tags of the cement that had flown into the tubules (Fig. 9). Etching with 30 % phosphoric acid seemed to create a further widening of the tubules at their opening (Fig. 10). A comparable effect was observed for the citric acid, but the tubules seemed to be filled with a homogenous substance after this treatment, different from the debris (Fig. 11).

Washing with Tubulicid removed parts of the debris but dit not open the tubules

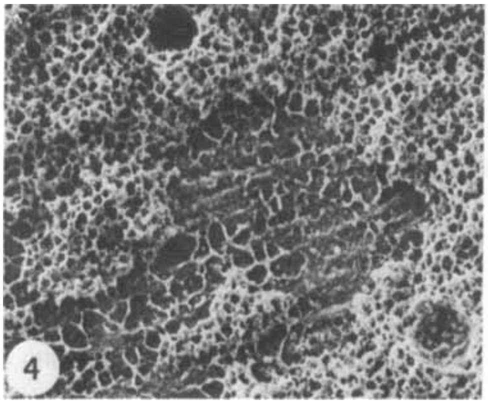

Fig. 4. Polished dentin with adhering polycarboxylate cement. x 500.

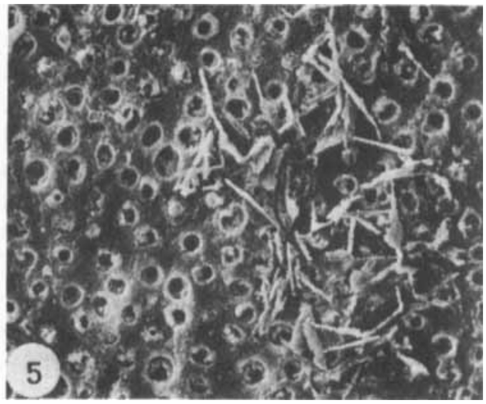

Fig. 5. Etched dentin with small increments of adhering polycarboxylate cement. x 1000.

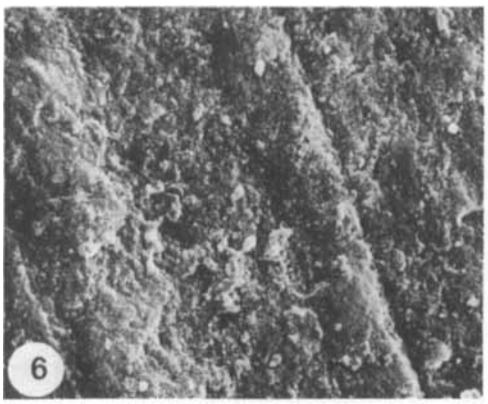

Fig. 6. Dentin polished on paper 800 and rinsed with water. x 3000.

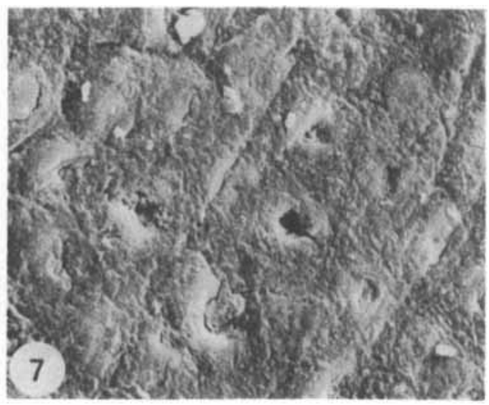

Fig. 7. Dentin polished and brushed with pumice. x 3000.

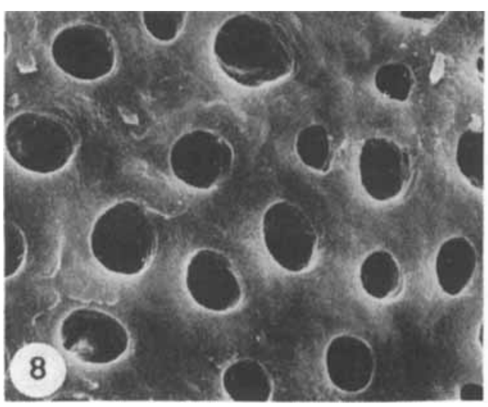

Fig. 8. Dentin polished and etched with H_3PO_4, 1 % for 10 sec. x 3000.

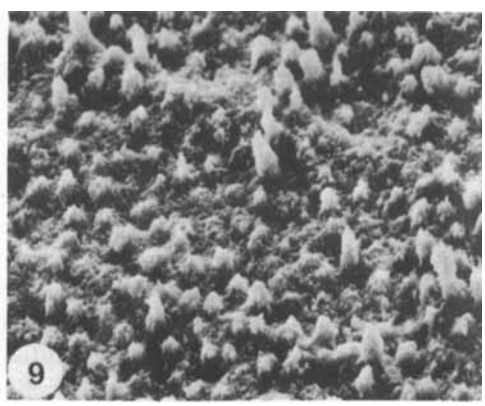

Fig. 9. Composite resin cement after removal from etched dentin. x 1000.

(fig. 12). Etching the surfaces with zinc phosphate cement liquid (Fig. 13) and the polycarboxylate cement liquid (Fig. 14) also seemed to remove the debris layer. The measurement of surface roughness showed that untreated, brushed and etched surfaces had an arithmetical mean roughness (R_a) in the range of 0.3–0.7 μm. The maximal roughness values (R_t) were in the range of 2–5 μm.

Significant differences in the surface roughness due to variations in surface treatment were not found.

DISCUSSION

The present study showed that the bond strength to smooth dentin varied considerably among the different cements. The values obtained for zinc phosphate cement were somewhat higher than those previously reported by Smith (17), and about twice that reported by Chan et al. (7). The latter study, however, used dentin surfaces which were rougher than those used in the present study, and it has been shown that increased surface roughness can lead to both larger air entrapment and a concentration of air bubbles in the cement close to the interface (9). This may explain the noted difference in bond strength. Swartz and Phillips (18) found no adhesion between zinc phosphate cement and wet dentin, but the preparation of the dentin was not comparable to that of the present study.

The observed bond strength of polycarboxylate cement to untreated dentin compares well with that previously reported (3, 16, 17). It is, however, more than twice the values reported by Chan et al. (7). A similar difference was found for the EBA cement, and again the difference in surface structure could explain the differences in bond strength. The composite resin cement had no measurable bond to untreated dentin in the present study. This is in agreement with what has been found for other composite materials (8, 16).

The present study also showed that the bond strength of all cements was markedly influenced by the surface structure of the dentin. A polished surface that was sprayed with water only, seemed to provide the best possibilities for the establishment of a bond between the dentin and zinc phosphate or polycarboxylate cements. The inspection in SEM showed that such surfaces were covered with a layer of grinding debris. The various treatments which reduced or covered this debris had a decreasing effect on the bond strength of zinc phosphate and polycarboxylate cements. This is in agreement with the results published in earlier studies (8, 15, 16). It has been shown that the polycarboxylate cement is bonded to hard dental tissues by an interaction between calcium ions and the carboxyl groups of the polyacrylic acid (1). An explanation for the reduction of bond strength could be that the dentinal surface consists of an aqueous-organic layer after etching (12), which would impede the establishment of a bond between the cement and the mineral components of the dentin. One may assume that the bond between zinc phosphate cement and dentin also relates to an interaction between the mineral substance and some parts of the cement, possibly the phosphate groups.

Another explanation for the decrease of bond strength might be that etched dentin surfaces were more wet than unetched because the moisture was free to leak out of the opened tubules (5). However, the fact that surfaces which were brushed with pumice also showed a reduced bond strength, could indicate that the debris layer which was dissolved by the cement liquids, could act as an intermediate layer in the establishment of a bond to dentin.

A pretreatment with phosphoric acid could obviously not promote the establishment of a bond between composite resin cement and dentin. This is in agreement with

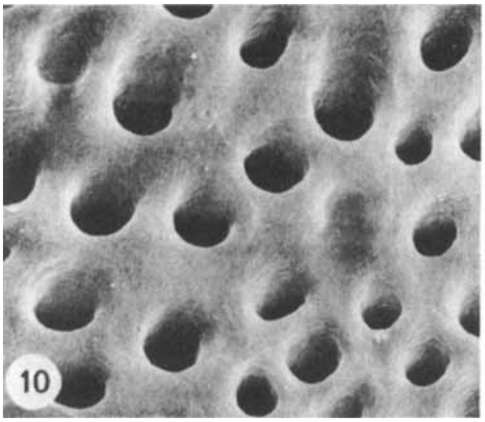

Fig. 10. Dentin polished and etched with H_3PO_4, 30 % for 60 sec. x 3000.

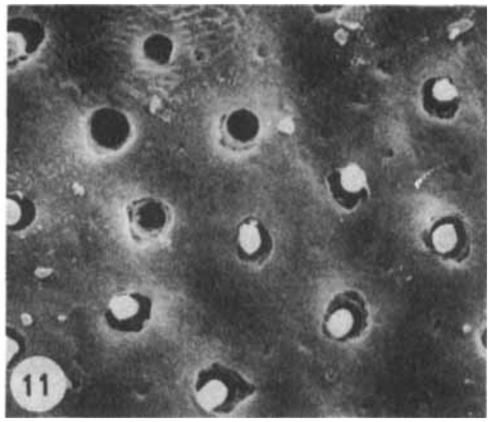

Fig. 11. Dentin polished and treated with a cavity cleanser containing citric acid for 60 sec. x 3000.

Fig. 12. Dentin polished and treated with Tubulicid® for 60 sec. x 3000.

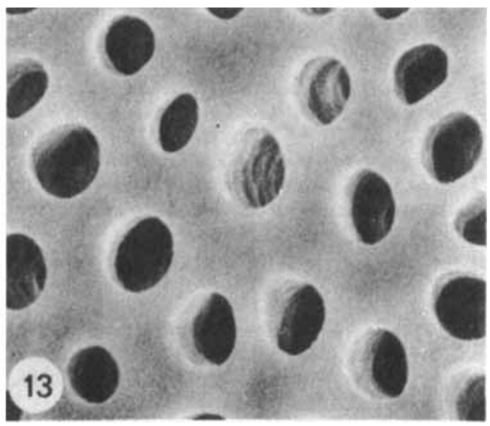

Fig. 13. Polished dentin after application of phosphate cement liquid for 60 sec. x 3000.

earlier studies (16). Etching with citric acid, however, resulted in a bond strength for the resin cement near to that of the polycarboxylate cement. An explanation for this difference could be that the tubules were open after etching with phosphoric acid but seemed to be filled with a solid substance after etching with the citric acid. As mentioned above, this could create a difference in the amount of water adhering to the surface, which again might influence the wettability of the dentin (5, 11).

An application of varnish or liners also reduced the bond strength of zinc phosphate, polycarboxylate and EBA cements.

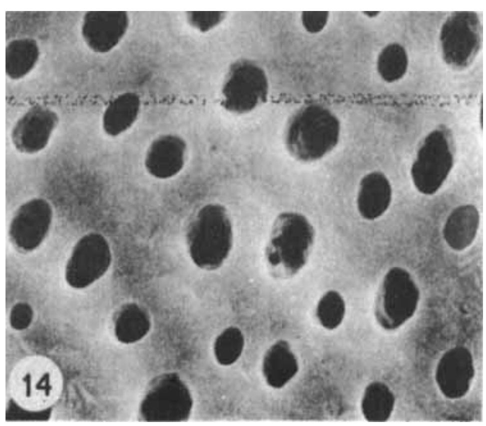

Fig. 14. Polished dentin after application of polycarboxylate cement liquid for 60 sec. x 3000.

These findings were in agreement with that previously reported (7). An explanation might be that the contact to dentin is prevented and that the adhesion to the liner or between liner and dentin was poor. Cyanoacrylates have been shown to have a good bond to dentin when used as a cementing medium for polymerized composites (2). When used as a varnish in the present study, the bonding values were approximately 10 times lower. Apart from the differences in materials and technique, a possibility exists that the cyano-acrylate film or parts of it was dissolved by the resin monomer.

REFERENCES

1. Beech, D.R. A spectroscopic study of the interaction between human tooth enamel and polyacrylic acid (polycarboxylate cement). Archives Oral Biol. 1972, 17, 907–911
2. Beech, D.R. Bonding of alkyl 2-cyanoacrylate to human dentine and enamel. J. Dent. Res. 1972, 51, 1438–1442
3. Beech, D.R. Improvement in the adhesion of polyacrylate cements to human dentine. Brit. dent. J. 1973, 135, 442–445
4. Brauer, G.M. Adhesion and adhesives, Pp. 49–96, in von Fraunhofer J.A., ed. Scientific Aspects of Dental Materials, Butterworths, London, England, 1975, pp. 471
5. Brännström, M. & Johnson, G. Effects of various conditioners and cleaning agents on prepared dentine surfaces: A scanning electron microscopic investigation. J. Prosthet. Dent. 1974, 31, 422–430
6. Buonocore, M.G. Preventive Dental Materials, Pp. 136–155 in Craig, R.G., ed. Dental Materials Review, University of Michigan, School of Dentistry, Ann Arbor, Michigan, 1977, pp. 287
7. Chan, K.C., Svare, C.W. & Horton, D.J. The effect of varnish on dentinal bonding strength of five dental cements. J. Prosthet. Dent. 1976, 403–406
8. Eden, G.T., Craig, R.G. & Peyton, F.A. Evaluation of a tensile test for direct filling resins. J. Dent. Res. 1970, 49, 428–434
9. Eick, J.D., Johnson, L.N., Fromer, J.R., Good, R.J. & Neumann, A.W. Surface topography: Its influence on wetting and adhesion in a dental adhesive system. J. Dent. Res. 1972, 51, 780–788
10. Eriksen, H.M. Pulpal response of monkeys to a composite resin cement. J. Dent. Res. 1974, 53, 565–570
11. Glantz, P.-O. On wettability and adhesiveness. Odontol. Revy, 1969, 20, supplement 17
12. Hoppenbrouwers, P.M.M., Driessens, F.C.M. & Stadhouders, A.M. Morphology, composition, and wetting of dentinal cavity walls. J. Dent. Res. 1974, 53, 1255–1262
13. Lee, H. & Swartz, M.L. Evaluation of a composite resin crown and bridge luting agent. J. Dent. Res. 1972, 51, 756–766
14. Myers, C.L., Ryge, G., Heyde, J.B. & Glenn, J.F. In vivo test for bonding strength. J. Dent. Res. 1963, 42, 907–911
15. Phillips, R.W., Swartz, M.L. & Rhodes, B. An evaluation of a carboxylate adhesive cement. J. Am. Dent. Assoc. 1970, 81, 1353–1359
16. Rider, M., Tanner, A.N. & Kenny, B. Investigation of adhesive properties of dental composite materials using an improved tensile test procedure and scanning electron microscopy. J. Dent. Res. 1977, 56, 368–378
17. Smith, D.C. A review of the zinc polycarboxylate cements. J. Canad. Dent. Assoc. 1971, 1, 22–29
18. Swartz, M.L. & Phillips, R.A. A method of measuring the adhesive characteristics of dental cements. J. Am. Dent. Assoc. 1955, 50, 172–177