Adherence of resin-based luting agents assessed by the energy of fracture

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The present study assessed the adherence of nine commercial resin cements by means of the wedge test. The beams of the test were made of a Ni-Cr-Be alloy, and the adhering surfaces were sandblasted with 250 μm Al_2O_3. The energy of fracture of the investigated cements varied from less than 10 J/m^2 to 121 J/m^2. The fracture energies were not influenced by 1) the thickness of the joint, 2) the width of the beams, 3) the use of an intermediary noncomposite resin, or 4) storage in water beyond 24 h. In spite of a rather high variability within groups, it is concluded that the wedge test may give relevant data on the performance of adhesive joints. □ *Adhesion; dental cements; dental materials*

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The retention of porcelain or composite inlays/onlays is normally provided by resin-based luting agents. As the name indicates, resin-bonded bridges are also cemented with luting agents of this type. A considerable number of procedures have been devised to make the inner surfaces of restorations amenable to resin-bonding: electrolytic etching, sandblasting, and silanization, just to name a few.

To evaluate the efficacy of a surface treatment or a resin cement, many different test methods have been used. The tests include shear tests and tensile tests (1) (or what one would like to believe are shear and tensile tests). The test specimens may be composed of the resin cement in the form of a cylinder, applied to one surface of the substrate (2). In other tests the resin cement is present in a thin layer between two bonded surfaces (3). A review of the literature shows, however, that results obtained with the same combination of materials, but in different laboratories, may differ widely. For example, using the resin cements Superbond and Panavia EX and the base-metal alloy NP2, one study (4) reported bond strengths of 22 and 18 MPa, respectively, whereas another study (5) found 26 and 44 MPa, respectively. In the same manner, one study (6) obtained bond strengths to the Co-Cr alloy Vitallium of 5 MPa after sandblasting and 19 MPa after electrolytic etching, whereas comparative values measured in another study (7) were 40 and 23 MPa, respectively. There may, of course, be many explanations of this erratic behavior; one of these is that it is not the bond itself that is being tested but rather an unknown combination of mechanical properties and shape factors. This possibility has been discussed by van Noort et al. (8, 9) in the case of dentin adhesives. Another problem with the conventional methods for measuring bond strengths is the relatively high standard deviation of the measured values: coefficients of variation of more than 30% are a common finding (4, 7). This makes valid comparisons between systems difficult and impedes further developments in the field.

In another approach to the assessment of bonding, not the bond strengths but the bond energies are measured (10). The method is adapted from the aviation industry and is particularly well suited to investigate the performance of an adhesive bond exposed to a hostile environment (11). The procedure for measuring bond energies is named 'the wedge test' or 'the double cantilever beam test' (DCB test) and has so far, with one exception, not been used in dental research (12).

It was the aim of the present study to measure the adherence of several commercial luting cements to the surface of a nonprecious alloy. The adherence was assessed by means of the wedge test, and the measurements served as a basis for an analysis of the possible role of some factors not considered in the mechanics of the wedge test (11).

Materials and methods

The wedge (or DCB) test is a cleavage test. It has been described in detail elsewhere (11), and only a brief review of its principle will be given here. The test makes use of a pair of identical beams that are glued together with the adhesive to be investigated. After hardening of the adhesive, a wedge is introduced in the joint, as illustrated in Fig. 1, to create a fissure. A known stress is produced by elastic deformation of the beams through the introduction of the wedge. The fissure propagates while elastic energy stored in the beams is released, and two new surfaces are created. If the sample is long enough, the crack propagation will stop when the release rate of elastic energy is equal to the energy necessary to form a new surface of unit dimensions. After approximately 24 h in water at 37°C, the fissure has reached its full length (11). The length (l) of the fissure is measured, and the energy of fracture (R) is calculated as $R = (3E/16)(t^3d^2/l^4)$, where t is the thickness of the beams, d the separation of the beams caused by the wedge, and E the modulus of elasticity of the beams. It is important to notice that for a given geometry and for a wedge of given dimensions, the energy of fracture depends only on E and on the length of the fissure at equilibrium. Moreover, the crack propagation under water is a good simulation of clinical fracture conditions.

The luting cements used in the study are listed in Table 1. ABC, INF, LC, PDC, and TL are dual-curing cements, whereas COM, DL, PEX, and SB are chemically curing. Only the adhesion stemming from the chemical cure of the cements was assessed. According to information given by the manufacturers, all resin cements except INF, PEX, and SB are based on 'normal' dimethacrylates without special adhesive properties. INF, PEX, and SB, on the other hand, contain monomers specially designed for adhesion. The beams were made of Rexilium III (Jeneric-Symphyse, USA), an alloy containing Ni, Cr, Mo, and Be as its main components and with a modulus of elasticity (E) of 220 GPa (12). The beams were approximately 45 mm long; the thickness and width will be described below. The beams were sandblasted with Al_2O_3 with a grain size designation of 250 µm and at a pressure of 0.4 MPa. An inspection showed that the sandblasting introduced a strain in the treated surface, resulting in a slight curvature of the originally plane specimens. For this reason both sides of the specimens were sandblasted, to produce plane specimens. (The distortion caused by sandblasting seems to be a hitherto unheeded source of inaccuracy.)

By means of double-sided adhesive tape, the two beams of a pair were affixed in a parallel and aligned manner to the two arms

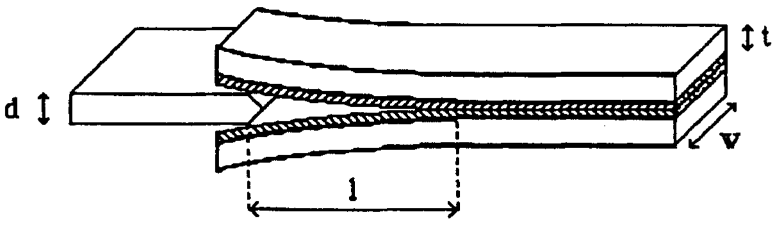

Fig. 1. Schematic representation of the wedge test. The wedge is introduced into the adhesive joint, causing a separation (d) of the beams. The length of the fissure is l; the thickness of the beams is t; and the width of the beams is w.

Table 1. List of resin cements used in the study

Code	Name	Manufacturer	Batch no.
ABC	ABC Dual	Vivadent, Lichtenstein	Base: 460057 Cat.: 460058
COM	Comspan Opaque Resin	Caulk Dentsply, USA	Base: 9206251 Cat.: 920625 Base: 9205191 Cat.: 920519
DL	Duralingual	Unitek Corp., USA	Primer: 920228 Composite: 920313
INF	Infinity	DenMat, USA	Paste: 811039 Cat.: 809048
LC	Luting cement	3M Co., USA	A Clear: 2184 B03 B Clear: 2177 B02
PEX	Panavia EX	Kuraray Co., Ltd, Japan	Powder: DN-706 Liquid: 2078
PDC	Porcelite Dual Cure	Kerr, USA	Base Univ.: 24323 Cat.: 27213
SB	Super Bond	Sun Medical Co., Ltd, Japan	Cat.: 101012 Monomer: 20603 Pol. Clear: 20401
TL	Twinlook	Kulzer, Germany	Base: 94.06.027 Cat.: 93.05.026

of a small vise. The luting cements were mixed in accordance with the instructions of the respective manufacturers and applied to the free side of both beams. The arms of the vise were pressed together, and the bonded specimen left to harden for 10 min at room temperature. The specimen was then separated from the vise and placed at 37 °C in a thermostated oven. After 24 h the excess of cement was removed by polishing the sides of the beams under water on carborundum paper nos. 800 and 1200, exposing the joint in its full length on both sides of the specimen. A steel wedge 460 μm thick was introduced in the joint by means of a specially designed bench with two perpendicular micrometer screws to direct the movements of wedge and joined beams. The separated DCB was then stored in water at 37 °C for another 24 h (except in one series, in which the storage time in water was 48 h). To inhibit corrosion of the knife edge, 0.1% of K_2CrO_4 was added to the water (13).

After the specified time in water the specimen was blown dry and placed under a stereomicroscope at a magnification of 4 × 10. The length (l) of the fissure (Fig. 1) was marked with a very fine felt point and measured with a ruler to the nearest 0.1 mm. The length of the fissure was determined on both sides of the specimen, and the mean value calculated.

The measurement of the separation (d) (Fig. 1) was carried out in a microscope at a magnification of 10 × 10. Insertion in the above formula of t, l, and d gave the energy of fracture R.

To investigate the possible influence of variables other than those entering the formula, we also measured the thickness of the joint and the width of the beams at the place marked as the end of the fissure. This was done in a microscope with measuring ocular at a magnification of 40 × 10 and with a micrometer screw, respectively. For each set of experimental conditions five sets of paired beams were investigated.

After the measurements the resin cement was eliminated by combustion at 600 °C for 30 min, followed by scrubbing with a brush under running water. The beams were then sandblasted anew, as described above, and ultrasonically cleaned in water. After a renewed measurement of the thickness of the beams, the specimens were rinsed with ethanol, dried with a hair dryer, and were now ready for a new series of experiments.

The adherence of nine commercial resin

Table 2. Experimental conditions and energy of fracture of the investigated resin cements (mean ± SD)

Resin cement*	Time in water (h)	Thickness of beam (mm)	Thickness of joint (μm)	Width of beam (mm)	Energy of fracture (J/m²)	Coefficient of variation (%)
ABC−	24	1.31 ± 0.10	46.8 ± 9.1	3.86 ± 0.32	38.0 ± 5.8	15.2
COM−	24	1.33 ± 0.10	40.2 ± 8.4	4.42 ± 0.39	35.0 ± 11.4	32.6
COM−	24	1.31 ± 0.10	185.2 ± 67.2	3.96 ± 0.35	31.1 ± 6.2	19.9
COM+	24	1.32 ± 0.10	33.4 ± 6.2	4.27 ± 0.40	35.9 ± 7.8	21.7
COM−	48	1.31 ± 0.10	174.0 ± 43.2	4.09 ± 0.39	28.4 ± 1.6	5.6
DL+	24	1.36 ± 0.10	107.8 ± 20.5	4.93 ± 0.39	32.0 ± 6.5	20.3
INF−	24	1.72 ± 0.04	154.2 ± 55.7	5.75 ± 0.10	83.8 ± 22.4	26.8
LC−	24	1.72 ± 0.04	109.0 ± 43.4	5.84 ± 0.09	<12.3	
PEX−	24	1.34 ± 0.10	43.2 ± 6.8	4.79 ± 0.41	45.5 ± 14.7	32.3
PDC−	24	1.33 ± 0.10	40.2 ± 6.0	4.51 ± 0.41	38.3 ± 11.4	29.8
SB+	24	1.35 ± 0.10	53.2 ± 8.3	4.85 ± 0.40	121.4 ± 35.0	28.8
TL−	24	1.33 ± 0.10	54.2 ± 11.8	4.59 ± 0.41	24.0 ± 3.8	15.8

* The use of an intermediary, noncomposite resin is indicated with a +.

cements was determined. In three additional series, we investigated the influence of 1) an adhesive joint of extraordinary thickness, 2) an intermediary, noncomposite resin, and 3) water storage for 48 h (see Table 2).

The statistical methods used were analysis of variance and four-dimensional regression analysis (14). Bartlett's test (14) was used to investigate the underlying assumption of identical standard deviations of the measured energies of fracture. As the standard deviations could not be assumed to be the same for all groups, a transformation was performed. A suitable transformation of the calculated fracture energies was obtained by using ln R as variable (14). The application of Bartlett's test (14) showed that the standard deviations of the transformed variable may be considered identical, enabling the use of analysis of variance and regression analysis as statistical tools (14).

Results

As was seen in the stereomicroscope, the fracture accompanying the development of a fissure was in all cases except INF of the adhesive type—that is, located between resin cement and metal surface. Occasionally, the adhesive failure shifted from the surface of one beam to the surface of the other beam, as illustrated in Fig. 2. With the resin cement INF, the main part of the fissure was located inside the cement, and the fracture was thus of the cohesive type. In some specimens the separation (d) of the beams was somewhat larger than the thickness of the wedge, because fragments of crushed resin cement were squeezed between the beams together with the wedge.

The measured variables and the energies of fracture are presented in Table 2, together with the coefficients of variation. With the resin cement LC, the fracture energy was so low that four of five pairs of beams separated completely on insertion of the wedge. No

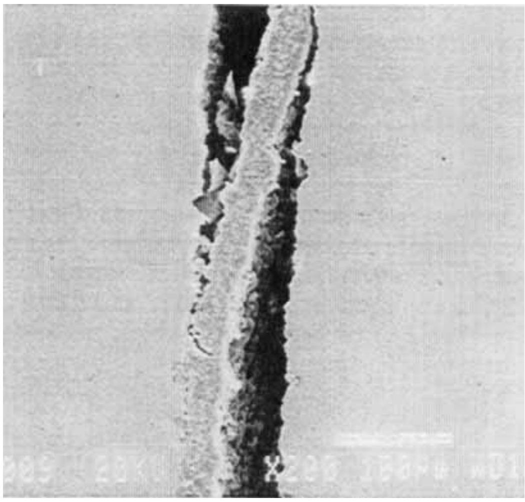

Fig. 2. Fissure between the two beams in a wedge test. The luting cement is seen between the beams. In the case shown, the fracture shifted from the surface of one beam to the surface of the other beam.

further statistical treatment was performed with this group. The statistical analysis showed that SB gave higher fracture energies than INF ($p < 0.05$), that INF gave higher fracture energies than PEX ($p < 0.001$), and that PEX gave higher fracture energies than the other cements ($p < 0.05$). When SB, PEX, INF, and LC were excluded, no difference was found between the remaining nine groups of Table 2 ($p > 0.05$).

The possible influence of beam dimensions (thickness and width) and joint thickness was analyzed by means of the transformed variable $\ln R$ in the following manner. Disregarding group LC and combining the COM-, 24 h, groups, with small and extraordinary thickness of the joint (see Table 2), 10 groups were formed. For each variable, and within these 10 groups, the deviations between mean value and individual values were calculated. Taking the deviations in $\ln \bar{R}$ as the dependent variable, and deviations in beam dimensions and joint thickness as independent variables, four-dimensional regression analysis was performed. The analysis failed to show with statistical significance the influence of the independent variables.

As an estimate of the accuracy of reading in the determination of the length of the fissures, the difference between the fissure length determined on the two sides of each specimen was calculated. From the 55 duplicate determinations of fissure length, the standard deviation of a reading was computed as 0.6 mm (14). Hence, for each specimen the fissure length (a mean value of two measurements) is obtained with a standard deviation of $0.6/\sqrt{2} = 0.4$ mm.

Discussion

In the present study the DCB test was utilized in the field of dental materials, and the possible influence of several experimental variables was examined. The fracture energies were found to vary among the investigated resin cements, and SB, INF, and PEX performed better than the other cements. A physicochemical explanation of this superior performance cannot be given at the moment, but according to the manufacturers, SB, INF, and PEX do contain monomers that are specially designed for adhesion.

The fracture energies obtained with SB, PEX, and DL are in good agreement with earlier findings (12). It is noteworthy, however, that the fracture energies of the cements presented in Table 2 in many cases are not well correlated with data on bond strength (1, 4, 5, 12, 15, 16). This indicates that bond energies and bond strengths are two distinct properties of a resin cement, and one may ask which of the two values provides the better measure of clinical predictability. To answer this question, long-term clinical studies are necessary, but at present only very few reports exist that permit the distinction between performance of resin cements. In one study, restorations cemented with PEX showed less failure than 'normal' resin cements (17), and in another study the retention provided by SB was superior to the retention of the 'normal' cements (M. Degrange. Unpublished observations). A comparison with Table 2 will show that such clinical data support the clinical relevance of the wedge test.

The finding that the thickness of the joint does not affect the measured fracture energies is in agreement with earlier data (11), and this is also the case with the lack of influence of prolonged water storage of the joint (11). No difference could be demonstrated between fracture energies with and without the application of an intermediary, noncomposite resin. This may be explained by the relatively low viscosity of the resin cement in itself, allowing a complete penetration of the cement into the irregularities of the sandblasted metal surface.

An undesirable feature of the wedge test—at least under the conditions utilized in the present study—is the relatively high standard deviation. The coefficients of variation (Table 2) vary between about 6% and 33%, with a mean value of 23%. Although this may not be higher than what is found in other studies on adhesion (4, 7), it does complicate research, focusing on the development of more reliable resin cements. In an attempt to analyze the sources of variation, we may write

$$\left(\frac{\Delta R}{R}\right)^2 = 16\left(\frac{\Delta l}{l}\right)^2 + 9\left(\frac{\Delta t}{t}\right)^2 + 4\left(\frac{\Delta d}{d}\right)^2.$$

where Δ is a measure of the inaccuracy of the associated variable (14). Assuming as typical values: $\Delta l = 0.4$ mm, $l = 28$ mm, $\Delta t = 0.01$ mm, $t = 1.3$ mm, $\Delta d = 10$ µm, and $d = 450$ µm, we obtain $(\Delta R/R)^2 = 0.00327 + 0.00053 + 0.00198$, or $(\Delta R/R) \approx 8\%$.

It appears that, in particular, a precise determination of the length of the fissure is important. It may be argued that the true fissure length cannot be determined in a stereomicroscope at a magnification of 4 × 10. However, this is true at any magnification, and it is quite possible that the use of a more powerful microscope will give the same degree of variation in the determination of fissure length. Further, it should be noted that the inaccuracy of 0.4 mm estimated above is a maximum value, since inhomogeneities in substrate and adhesive may in fact give a fissure with different lengths at the two sides of the joined beams.

The contribution to the inaccuracy of R stemming from the variables l, t and d was estimated above as approximately 8% of R, whereas the mean coefficient of variation was found to be 23%. Another source of variation is the sandblasting, which in spite of a regular appearance may not be uniform between specimens. The bending of the beams mentioned earlier may also give rise to a variation in the results. Although sandblasting on both sides of each beam tended to reduce this problem, some stresses may be induced in the joint before the insertion of the wedge. This may be particularly true in the present work, in which the resin cements polymerized between beams assembled in a vise. After loosening of the vise, any bending stress in the beams will be transferred to the adhesive joint. A solution to the problem would be to use a guiding device, by means of which the resin cements are permitted to polymerize between unstrained beams.

To conclude, the present work has investigated the use of the wedge test in the assessment of the adherence of resin-based luting agents. In spite of rather high variability, the test may give relevant data on the performance of adhesive joints. Resin cements with monomers specially designed for adhesion performed better than cements without such a feature.

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