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Porous ceramic lamellae for orthodontic ceramic brackets

Part I *Fabrication and characterization*

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This study was undertaken to test a new and original orthodontic bracket base, consisting of a porous lamella, which was designed to facilitate removal of ceramic brackets from the enamel surface after treatment. In the phase of the study presented here, porous pellets were made by bonding coarse alumina particles (calcined or fused) with mullite, formed by their reaction with fine quartz particles during firing of the mixture at 1700 °C. After machining the pellets to the desired shape, nine types of lamellae with different porosities were attached to the brackets using two different adhesive resins, and also bonded to bovine enamel using the same adhesives. The tensile bond strengths for the assemblies were determined so that the lamellae and the bonding adhesive that might be suitable for clinical application could be selected for testing *in vitro* in the second phase of the study.

1. Introduction

Tooth movement in orthodontic treatment is frequently carried out by bonding a bracket firmly to the tooth surface with a dental resin and placing an arch wire in the bracket slot which, under tension, applies a gentle force to the tooth thus causing it to be moved in the desired direction.

Brackets used for this purpose are generally made of stainless steel because of its mechanical strength and physiological inactivity. However, the appearance of such brackets is a distinct disadvantage, as they are very noticeable. This drawback resulted in a search for a bracket that combines strength with a more pleasant appearance.

This search led to the use of ceramics, which are translucent or transparent while also being hard and strong. Most of the currently available ceramic brackets are composed of aluminium oxide, either in monocrystalline or, more usually, polycrystalline form. "Single crystal technology" sapphire brackets are manufactured from man-made, single-crystal alumina and are optically clear. Polycrystalline ceramic brackets are translucent and satisfactorily match tooth colour [1].

Ceramic brackets may be bonded to the enamel surface using one of three retentive methods: (a) chemical mediator (usually a silane coupling agent) between the bracket base and the adhesive; (b) mechanical retention via indentations and/or undercuts in the bracket base; or (c) a combination of these. Chemically-retained ceramic brackets have a smooth surface coated with a layer of silica glass. This is painted with silane to promote bonding to the adhesive resin [1, 2].

These methods have produced exceptional bond strengths [3–5]. However, high bond strengths, the low fracture toughness of the ceramics and the method employed for debonding have caused complications during bracket removal at the end of orthodontic treatment [6–10]. The main complications associated with ceramic bracket debonding can be listed as follows:

(a) *Bracket fracture*. When debonding a metal bracket, its malleability permits it to be peeled from the tooth surface. Ceramic brackets, being rigid and brittle, often fracture upon debonding [6–8]. In such a case, a significant risk for the patient is the accidental ingestion or aspiration of bracket fragments, which because of their radiolucency may not be detectable on radiographs. Also, fragments may cause damage to oral soft tissue and possible eye injury to the patient, clinician or assistant.

(b) *Enamel fracture*. Because of the high bond strength and the inability of ceramic brackets to flex, greater forces are usually required to remove them during debonding, resulting in a higher incidence of enamel damage during debonding [6–10].

(c) *Discomfort for patient while debonding ceramic brackets*. This is probably related to the high level and direction of the force applied during debonding, which sends a shock wave through the tooth that is both alarming and painful for the patient.

(d) *Removal of ceramic brackets by grinding*. When the debonding technique fails, leaving a section of the bracket still attached to the enamel, grinding the remaining ceramic becomes the option of choice to remove it. Grinding is usually conducted with high-speed diamond burrs or low-speed green stones. The procedure is time-consuming and despite the use of a water spray the heat which can be generated may

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affect the dental pulp and the subsequent vitality of the tooth [11]. It is also particularly difficult to differentiate the appearance of the retained ceramic and adhesive from the underlying enamel, thus risking inadvertent damage to the enamel.

Although new bracket base designs and alternative debonding techniques to the conventional mechanical methods have been introduced, including electrothermal, ultrasonic and laser debonding, clinicians still encounter problems and removal has been identified as a major disadvantage of the use of ceramic brackets [12–14].

It was considered that these problems might be solved by interposing a layer between the bracket and tooth composed of an open porous ceramic lamella whose pores were filled with the adhesive. The lamella was to consist of ceramic particles each having ceramic bonds to its neighbours which should be small in cross-sectional area. Such a layer might be crushed by the action of conventional pliers during bracket removal and yet, with the pores filled with adhesive, the bracket should be held firmly to the tooth (Fig. 1).

The aims of the study were, therefore, to fabricate porous lamellae with the required microstructure, and to test the effectiveness of lamellae when used with commercial brackets. In order to accomplish these aims the following steps were executed:

1. Porous lamellae having various porosities were fabricated using two different types of alumina starting powders.
2. Nine types of lamellae with different porosities, ranging from 27% to 50%, were attached to commercial brackets by means of mechanical adhesion, and the optimum porosity for the intended application was found by investigating the tensile bond strength of these bracket/lamella assemblies to enamel.
3. The lamellae chosen after step 2 were further investigated under simulated clinical conditions *in vitro* in order to ascertain their effectiveness and suitability for clinical use.

In the first phase of the study steps 1 and 2 were performed and are described here. In the second phase, step 3 was completed and the results of that work will be described in part II of the paper.

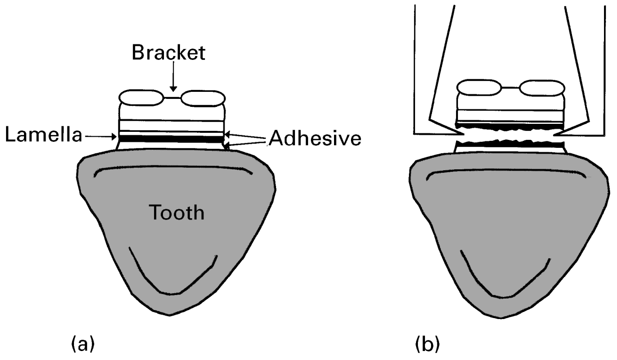


Figure 1 Diagrams of bracket/lamella assembly: (a) new bracket/lamella assembly bonded to tooth; (b) anticipated mode of failure during debonding with conventional pliers.

2. Materials and methods

After evaluating some biocompatible ceramics, including hydroxyapatite and calcium aluminate, it was decided that porous alumina would be most suitable for the lamellae because it possesses a number of desired properties, namely good aesthetic appearance, excellent corrosion resistance and biocompatibility. A range of alumina ceramics with different porosities were processed using an approach similar to one previously patented [15]. This involved forming mullite bonds between alumina particles by reacting the alumina with quartz.

2.1. Starting materials

Two coarse alumina powders were selected. These were a calcined α-alumina (purity 99 + %) and fused α-alumina both obtained from Aldrich (Dorset, England).

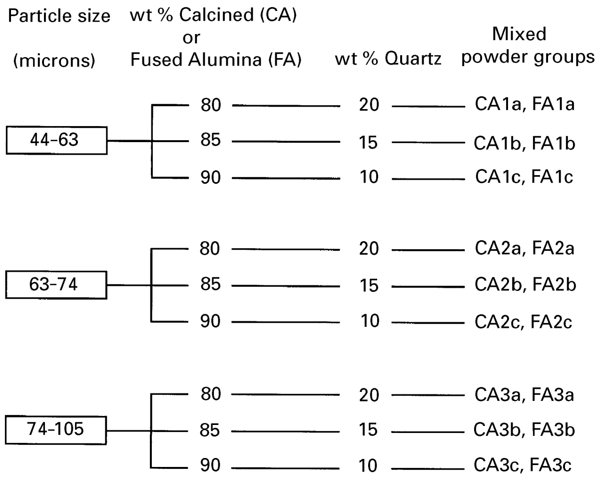
Both alumina powders had particles in the range $-100 + 325$ mesh (149–44 μm). For each type of powder, three batches having narrower size ranges were produced by sieving. The batches had size ranges of 44–63 μm, 63–74 μm and 74–105 μm.

In this study, quartz powder having a median particle size of 2.4 μm was used for all lamella groups to produce the permanent bonds.

The temporary binder used in this study was poly(ethylene glycol) (Carbowax, 20 M Union Carbide Corp., USA).

2.2. Processing and firing

The component powders were dried and then weighed out to an accuracy of ± 0.01 g to obtain batches of 10 g of the required composition. The calcined and fused alumina powders for each size range were mixed with quartz powder as the minor component in the proportions 90 : 10, 85 : 15 and 80 : 20. The initial mixing was performed by rolling the powders on a sheet of paper. This is not adequate because particles of different size readily segregate. The mixtures containing calcined or fused alumina were labelled as CA or FA respectively. Fig. 2 illustrates the mixtures produced.



Particle size (microns)	wt % Calcined (CA) or Fused Alumina (FA)	wt % Quartz	Mixed powder groups
44-63	80	20	CA1a, FA1a
	85	15	CA1b, FA1b
	90	10	CA1c, FA1c
63-74	80	20	CA2a, FA2a
	85	15	CA2b, FA2b
	90	10	CA2c, FA2c
74-105	80	20	CA3a, FA3a
	85	15	CA3b, FA3b
	90	10	CA3c, FA3c

Figure 2 Mixtures produced in this study.

12 g of binder was dissolved in 30 g of distilled water. A 10 g mixture of calcined alumina and quartz powder (CA) was poured into this binder solution and stirred for 20 min at a rate of 60 rpm. A thick paste formed upon stirring which prevented segregation of different particle sizes. This process was repeated for each CA mixture.

For the fused alumina and quartz groups (FA), the weights of binder and water were halved, as this alumina powder was not porous and consequently did not absorb any water. The resultant thick paste was stirred as before.

Samples of 5 g of these pastes were uniaxially pressed into pellets 25 mm in diameter using a cylindrical single-ended steel die. A compaction pressure of 40 MPa was used. Excess binder solution escaped during the pressing operation.

The pellets were left at room temperature for 2 days to dry in air before firing. After burning the binder out at 500 °C for 2 h, the pellets were placed in an alumina boat. They were then fired in air at 1700 °C for 1 h in an electrically heated kiln. The heating and cooling rates were the same, 5 °C per minute.

2.3. Characterization of the fired pellets

The microstructures and compositions of the fired pellets were examined using scanning electron microscopy (SEM), and X-ray diffraction (XRD), respectively. The true porosities were determined using image analysis.

A CAMSCAN Series 2A scanning electron microscope equipped with a Link An 10000 energy dispersive spectrometer (EDS) was used in the characterization of the fired materials. The specimens for SEM inspection were impregnated with epoxy resin and polished.

The X-ray diffraction analyses were carried out using CuK_α radiation in a Philips X-ray diffractometer (PW 1050/25 goniometer and PW 1730 generator) with 50 kV tube voltage and 30 mA current. A scan rate of 2°/min was used over a range of 10–85° for all samples.

The porosities of the fired pellets were measured using a computerized video image analyser. The analysis of each group was carried out by measuring and comparing the areas of the dark (porosity) and light (ceramic material) fields on five scanning electron micrographs taken from different sintered pellets of the same group.

2.4. Machining of lamellae

The alumina pellets were machined to the desired final shape by using a low-speed annular saw with a diamond impregnated blade (Impregnated Diamond Products Ltd). The pellets were mounted on a cutting board with dental wax and then sliced into 4 mm × 5 mm × 0.5 mm thin pieces on a Microslice 2 cutting machine (Malvern Ltd., England). The cutting speed and pressure of the microslicer were taken from the data given for high alumina ceramics elsewhere in the literature [16].

After slicing, all pieces were cleaned in an ultrasonic bath for one minute in order to remove any debris.

2.5. Optimizing porosity

After fabricating the lamellae, a study to measure bond strength was conducted in order to decide on the optimum porosity for this application and to reduce the sample size for further *in vitro* testing. This study included nine types of lamellae as representatives of different porosities and was performed using tensile testing, which was selected because of the lower bond strengths of orthodontic brackets in tensile testing compared with the results from shear testing [17–20].

First, five types from the FA group and four from the CA group were chosen as representatives of different porosity levels. These are given in Table I. Twenty lamellae of each type were attached to mesh-based metal brackets of the type used for upper central incisors (Forestadent Ltd., Pforzheim, Germany). Metal rather than ceramic brackets were employed at this stage for reasons of economy. Ten lamellae of each type were attached to brackets either using one of two chemically-cured orthodontic bonding adhesives. These were a no-mix, paste-liquid system (Eurobond, Hudson Ltd., Sheffield, UK), and a two-paste, highly-filled composite (Concise, 3M Dental Products, USA). For attaching lamellae to the brackets with Eurobond, the bonding initiator (liquid) was applied to the surfaces of both the lamella and bracket to be bonded, and Eurobond adhesive paste was sandwiched between these two surfaces. Then the bracket/lamella assembly was immediately placed under constant load of 400 g in a specially designed jig. With Concise the following steps were performed. The lamella surface was coated with a mixture of Concise enamel bond resin A and resin B. A mixture of the bonding paste A and paste B was immediately applied to the bracket base in a thin film and the bracket was positioned on the lamella before the application of a constant load of 400 g. A bracket/lamella assembly joined with either adhesive was allowed to cure for 5 min under load. It was removed from the jig and put aside for 15 min of bench cure. Then the lamella was trimmed with a dental burr so that it had a closely similar area to that of the base of the bracket. In this process adhesive that had exuded under pressure was also removed. The mean area of the trimmed lamellae was determined for

TABLE I Porosities of the lamellae used in the tensile test

Lamella	FA1c	FA3b	FA2b	FA2a	FA1a	CA3c	CA2b	CA2a	CA1a
Porosity (%)	27	30.5	35	37	39	42	45.5	47.5	49

ten assemblies using an image analyser. This was found to be 12.34 mm^2.

Following this preparation, the bracket/lamella assemblies were bonded to bovine incisor teeth in accordance with normal clinical methods using the same adhesives. Teeth bonded to bracket/lamella assemblies were then mounted in plastic cups filled with a low-temperature-setting mounting resin leaving only the labial surfaces of the teeth and the bracket/lamella assemblies exposed. During mounting, a special jig was used to ensure that each bracket/lamella assembly was placed at the centre of its plastic cup, with the lamella parallel to the mounting resin surface for accurate application of tensile force. After mounting, the samples were stored in distilled water at 37 °C for 24 h prior to testing.

The test for tensile bond strength was carried out using a Lloyd M 5K testing machine (Lloyd Instruments plc, Hampshire, England). Tension was applied to the samples via a jig described elsewhere [17]. A crosshead speed of 1 mm/min was maintained and the force at the bond failure was recorded in newtons. The bond strength was then calculated on the basis of the stress at failure divided by the mean bond area of 12.34 mm^2 and expressed in MPa.

The differences in bond strength were investigated statistically using an analysis of variance (ANOVA). Any differences revealed by this procedure were further investigated using a Tukey's Honesty Significant Difference (HSD) multiple range test with a 95% confidence interval ($p < 0.05$).

3. Results

3.1. Characteristics of the fired pellets

Typical microstructures are shown in Fig. 3a and 3b for two compositions, FA1b and CA2c, respectively.

Fig. 4 shows a polished section of FA3b on which energy dispersive analysis was performed. The necks that developed between the alumina particles were shown to contain aluminium and silicon, and the XRD spectra of the sintered pellets from different groups exhibited peaks corresponding with only those of alumina and mullite. This suggests that the heat treatment was sufficient to cause the quartz and alumina to react to bond the particles together with mullite ($3Al_2O_3$ $2SiO_2$). No quartz was detected by XRD, although small residual amounts may not have been registered using this technique.

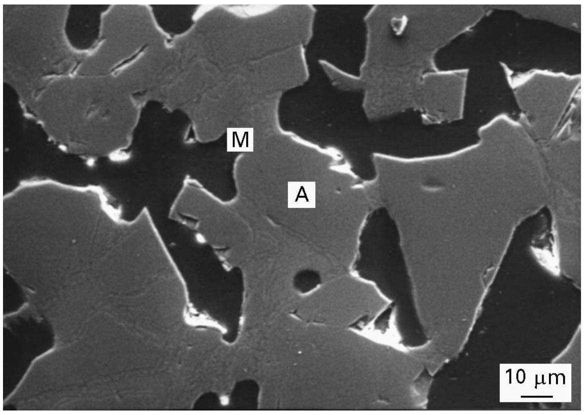


Figure 4 Scanning electron micrograph of a polished section of FA3b. EDS trace from M revealed the presence of Al and Si, and from A the presence of only Al.

From a comparison of the major peaks of the phases, it was seen that the formation of mullite increased with an increase in the added quartz content, as would be expected. Fig. 5 shows the relative peak intensities of the mullite (1 1 0) reflection, corresponding to a 0.539 nm d-spacing, and the α-alumina (1 1 6) reflection, which corresponds to a d-spacing of 0.1601 nm, for the mixtures of CA1a, CA1b and CA1c. This gives a semiquantitative measure of the mullite to the alumina ratio.

In Fig. 6, the true porosity is plotted against the added quartz content for each test group. It can clearly be seen that porosity decreased as the quartz content was increased.

3.2. Optimum porosity

The histogram in Fig. 7 illustrates that the assemblies had higher bond strengths when bonded with Concise adhesive than with Eurobond. The two-way analysis of variance, using bond strength as the dependent variable, demonstrated significant differences between the lamellae ($p < 0.001$) and between the two adhesives ($p < 0.001$). One-way analysis of variance and

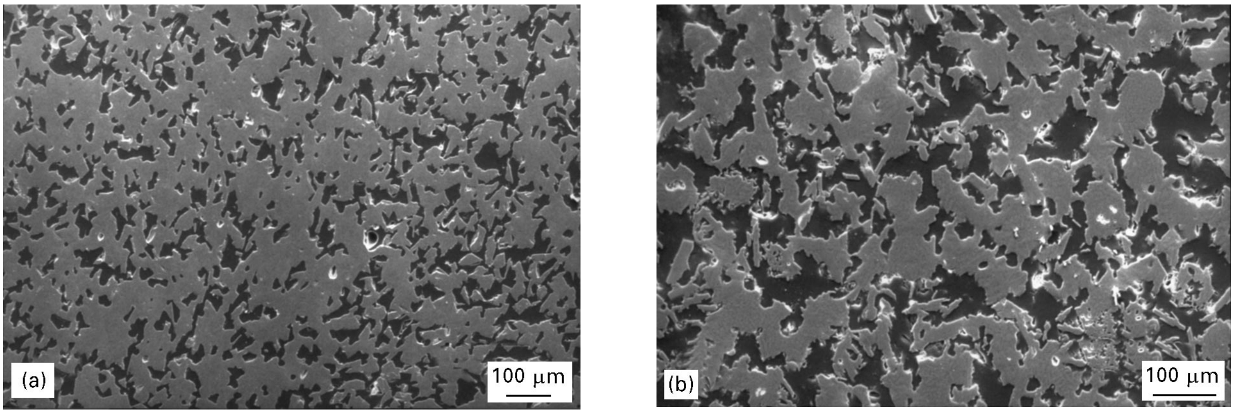


Figure 3 Scanning electron micrographs of polished sections of (a) FA1b, (b) CA2c.

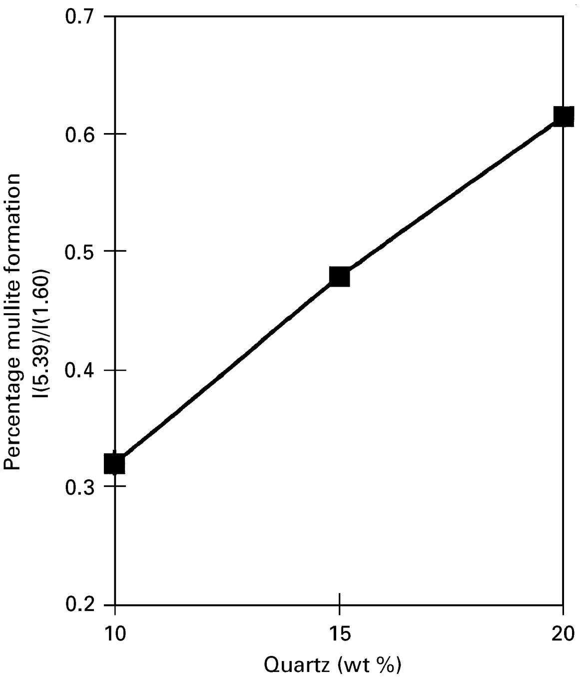


Figure 5 Relative peak intensities of mullite (1 1 0) to alumina (1 1 6) as a function of the added quartz.

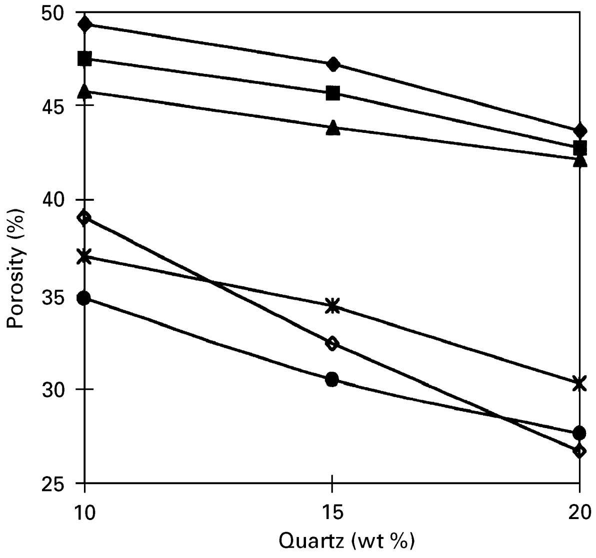


Figure 6 Porosities of the lamellae as a function of quartz content: ◆ CA1; ■ CA2; ▲ CA3; ◇ FA1; ∗ FA2; ● FA3.

the Tukey's HSD multiple range test indicated that the systems using CA2b and FA2a with Concise had the higher tensile bond strengths.

The CA2b (45% porosity) lamella had the highest mean tensile bond strength at 6.13 ± 1.79 MPa with Concise adhesive resin. The second highest bond strength was obtained by FA2a (37% porosity) with Concise (5.90 ± 1.70 MPa). CA2a (47% porosity) showed the highest standard deviation with both adhesive types, ± 1.91 and ± 1.90 MPa, respectively.

The predominant mode of bond failure for each group was at the bracket/lamella interface, in other words the brackets usually separated from the lamellae leaving the lamella on the enamel surface.

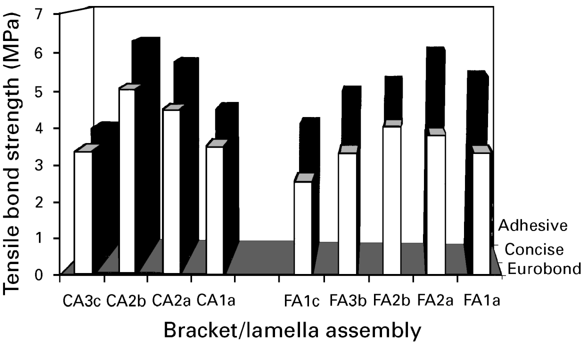


Figure 7 Tensile bond strengths of the lamellae tested.

4. Discussion

With the processing technique used to form the alumina lamellae, between 27% and 50% porosity could be produced. Although using spherical alumina particles would produce more regularly shaped and evenly distributed porosity, this type of powder is not thought to be commercially available for the particle sizes used in this study.

Since the sizes of the pores in the fired pellets were large, mercury porosimetry was not used to measure the open porosity. Insted, a computerized video image analyser was used in the study with micrographs taken from polished sections. The standard deviations of the porosity values were high for all groups because of the small sample sizes (five for each group), but the results were sufficiently accurate for the analysis to be statistically meaningful.

The pellets processed using the calcined alumina powders had the higher porosities (Fig. 6). This was because the calcined alumina starting powders were porous aggregates, while the fused alumina particles were dense and also because the former particles had a more irregular shape which adversely affected particle packing. During firing, the intraparticle pores in the aggregates were transformed into interparticle pores and thus the open porosities in the CA groups were increased.

The size of the alumina particles relative to those of the quartz affects how densely mixtures of the two pack. It would be expected that the greater the difference in size, the more densely the mixture would pack. After firing, it appears that this size effect on packing is retained for all cases except FA1a and FA1b as shown in Fig. 6.

No attempt was made to measure the mechanical properties of the lamellae as it was the complete system behaviour under simulated clinical conditions that was important.

The mean tensile bond strengths of the majority of the bracket/lamella assemblies constructed and bonded to the teeth with Concise adhesive exhibited clinically acceptable levels, being above the strength recommended by Reynolds of 4.9 MPa [21]. However, only one type of assembly with Eurobond adhesive had a mean bond strength value over this limit (CA2b).

It is apparent that the bracket/lamella assemblies bonded with Eurobond paste/liquid no-mix adhesive performed relatively poorly compared to those bonded with Concise, which is a highly-filled two-paste adhesive resin. In the majority of system failures with Eurobond, the adhesive resin stayed attached to the mesh base of the bracket leaving intact lamellae on the enamel surfaces. In the case of the Concise bonded systems, failure frequently involved partial fracture of lamellae. The difference in behaviour of the two adhesives may partly be explained by the fact that the Eurobond bonding initiator (liquid) enters the pores of the lamellae, which may adversely affect the blending of the two components and cause incomplete polymerization in the adhesive. In addition, the presence of initiator liquid in the pores may limit the entry of the adhesive paste. For the assemblies bonded with Concise, the pores were filled with a mixture of enamel bond resin. This is the same as the resin used in the highly filled paste employed to bond the lamella to the bracket.

Higher bond strengths with two-paste adhesive resins when compared to no-mix adhesive resins have been measured in several *in vitro* studies and it has been suggested that this was because the former contained smaller amounts of incompletely polymerized material [17, 22, 23].

Testing the lamellae with a wider range of orthodontic adhesives may result in different optimum porosities generating higher bond strength values. However, the present study suggests that the highest bond strengths would be likely to be obtained when both lamella groups are bonded with two-paste, chemically-cured, adhesive resins.

In the second part of this study, the lamellae of both types that exhibited the highest bond strength values with Concise adhesive, namely FA2a and CA2b, were evaluated with respect of their bond strengths and bond durabilities under simulated clinical conditions. Their bond failure sites and the force levels required for debonding with conventional pliers were also investigated as this is the proposed technique for the removal at the end of treatment.

5. Conclusions

The processing technique developed for the lamellae proved capable of producing porous mullite-bonded alumina ceramics with porosities from 27% to 50%.

For the CA group, the highest tensile bond strengths were achieved with 45% porosity for both types of adhesive resin used to bond them to brackets and bovine enamel. For the FA group, the highest bond strength with no-mix Eurobond adhesive was attained with 35% porosity, whereas for the Concise adhesive the highest value occurred for 37% porosity. The higher bond strengths were above the recommended minimum value [21].

References

1. M. L. SWARTZ, *J. Clin. Othod.* **22** (1988) 82.
2. S. E. BISHARA and T. S. TRULOVE, *Amer. J. Orthod.* **98** (1990) 145.
3. A. J. GWINNETT, *ibid.* **93** (1988) 346.
4. J. ØDEGAARD and D. SEGNER, *ibid.* **94** (1988) 201.
5. J. GHAFARI and S. S. CHEN, *J. Dent. Res.* **69** (1990) 1837.
6. E. R. STORM, *J. Clin. Othod.* **24** (1990) 91.
7. T. B. REDD and P. K. SHIVAPUJA, *J. Clin. Othod.* **25** (1991) 475.
8. S. L. GIBBS, *Br. J. Orthod.* **19** (1992) 191.
9. H. NEWESELY and B. ROSSIWALL, *Inf. Orthod. Kieferorthop.* **21** (1989) 577.
10. M. T. JEIROUDI, *Amer. J. Orthod.* **99** (1991) 97.
11. M. E. VUKOVICH, D. P. WOOD and T. D. DALEY, *Amer. J. Orthod.* **99** (1991) 505.
12. J. J. SHERIDAN, G. BRAWLEY, and J. HASTINGS, *ibid.* **89** (1986) 21.
13. K. STROBL, T. L. BAHNS, L. WILLHAM, S. E. BISHARA and W.C. STWALLEY, *ibid.* **101** (1992) 152.
14. D. B. BOYER, G. ENGELHARD and S. E. BISHARA, *ibid.* **108** (1995) 262.
15. H. NAKAJIMA, T. ITO and Y. MURAGUCHI, *USA Patent No*: *4,965,230* (1990).
16. G. W. FYNN and W. J. A. POWELL, In “Cutting and polishing optical and electronic materials” (Adam Hilger, Bristol, 1988) p. 3.
17. D. REGAN and R. VAN NOORT, *Eur. J. Orthod.* **11** (1989) 144.
18. A. J. OSTERTAG, V. B. DHURU, D. J. FERGUSON and R. A. MEYER, *Amer. J. Orthod.* **100** (1991) 251.
19. J. M. BORDEAUX, R. N. MOORE and M. D. BAGBY, *ibid.* **105** (1994) 552.
20. S. W. MERRILL, J. O. LARRY and C. B. HERMESCH, *ibid.* **106** (1994) 290.
21. I. R. REYNOLDS, *Br. J. Orthod.* **2** (1975) 171.
22. V. A. J. BUZZITTA, S. E. HALLGREN and J. M. POWERS, *Amer. J. Orthod.* **81** (1982) 87.
23. J. W. FERGUSON, M. J. F. READ and D. C. WATTS, *Eur. J. Orthod.* **6** (1984) 267.

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