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Adhesion of methacrylate-based composite materials to bone: a shear bond strength and scanning electron microscopy study

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The aim of the present study was an analysis of the adhesion of composite materials to fresh bovine bone tissue. The analysis was carried out by evaluating shear bond strength and scanning electron microscopy (SEM) results. All specimens showed a good composite–bone adaptation with strong binding between methacrylate-based materials and bovine bone. These composites could be used in an array of surgical fields in addition to fresh autogenous grafts.

1. Introduction

Considerable research has been focused on the tooth–composite interface and the achievement of a bonding to dentin [1–8]. The use of a dentinal bonding agent can improve the bond strength of composite to dentin, reducing microleakage and recurrent caries and facilitating the preparation of cavities without retentive undercuts.

The action of the dentin bonding agents is the creation of a bond to either the inorganic or organic portion of the dentin [5–9]. With the dentin adhesives of the second type it is necessary to pre-treat with EDTA to remove the smear layer and the inorganic plugs in the dentinal tubules [10–11]. One of the most efficient agents found to date is the Gluma Dentin Bond System (Bayer, Germany), including Gluma Cleanser (EDTA water solution).

Gluma Bond (a mixture of glutaraldehyde and 2-hydroxyethylmethacrylate) and Gluma Sealer (fluid resin) is described as an adhesive with a potential capability of bonding to the organic components of the dentin, such as collagen fibres: the bond strength between a restorative resin and dentin pre-treated with EDTA and Gluma is said to approach that of acid-etched enamel [12–14]. Other dentin adhesives, such as Scotchbond 2 Dental Adhesive System (3M, USA), interact primarily with inorganic dentinal components and show a preference for a surface which has not been subjected to complete smear layer removal. The smear layer must be prepared or primed by Scotchprep Dentin Primer, an aqueous solution of a hydrophilic methacrylate monomer (Hema) and an organic acid (maleic acid). Scotchbond 2 Light Cure Dental Adhesive, primarily composed of a hydrophilic methacrylate monomer, bis GMA, and a photoinitiator, wets out and penetrates the solubilized smear layer and, when polymerised, is locked into this substrate (6, 14–16).

The percentage of organic and inorganic materials is similar in dentin, radicular cement and bone tissue [7].

The aim of the present study is an evaluation of the shear bond strength of a posterior composite (P50; 3M, USA), used with two different dentin bonding systems (Gluma and Scotchbond 2) to fresh bovine bone tissue, and a scanning electron microscopy (SEM) analysis of the composite–bone interface.

2. Materials and methods

Seventy-six bone segments of bovine femur diaphysis were freshly harvested. The cortical surface was about 300 mm^2. The specimens were washed in saline solution and after manually removing the soft tissues and exposing a raw surface, were stored in saline solution at room temperature. After a short blast of oil-free compressed air the cortical bone of 30 specimens was treated for 60 s with a 0.5 M EDTA solution (Gluma Cleanser, Bayer, Germany): the treatment was carried out by gentle rubbing with a soaked cotton pellet, followed by water spray for 20 s and a blast of compressed air. Subsequently Gluma-Bond, a mixture of 5% glutaraldehyde and 35% 2-hydroxyethylmethacrylate (Gluma Bond, Bayer, Germany) was applied on the same sites for 60 s. Excess reagent was then removed by a gentle blast of oil-free compressed air and a thin layer of fluid resin (Gluma Sealer, Bayer, Germany) was placed with a cotton pellet.

The posterior composite (P 50) was applied in compliance with the manufacturer's instruction directly to the bone surface by means of a 8 mm × 4 mm Teflon tube and polymerized with a 3 M/LC (light curing) halogen lamp.

After air-drying, the bone surface of another 30 specimens was treated for 60 s with a hydroxyethyl-

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methacrylate-maleic acid aqueous solution (Scotchprep Dentin Primer, 3M, USA). After evaporation with a gentle blast of air, a fluid resin (Scotchbond 2 Light Cure Dental Adhesive, 3M, USA) was placed on the pre-treated bone and polymerized with a 3 M/LC (light curing) lamp.

Subsequently a posterior composite (P 50, 3M, USA) was applied using the previously described Teflon tubes, and polymerized with the same lamp.

All specimens were stored in saline solution and then mounted on special stabs for shear strength test on a universal testing machine, with a crosshead speed of 0.5 mm/min and was expressed as MPa. Another eight specimens were treated with the composite-bonding system Gluma/P50 as above: four of them were sectioned with a diamond burr and smoothed with a Sof-Lex Pon On XT disk (3M) for SEM observations at the bone–composite interface. The other four specimens were dehydrated with an ascending series of alcohol rinses, embedded in resin (Technovit 7200 VLC, Kulzer, Germany) and sectioned according to the Exakt cutting–grinding system [18] for observations under transmitted normal and polarized light microscopy. Another eight specimens were treated with the composite-bonding system Scotchbond 2 Dental Adhesive System/P50 as above: four of these specimens were sectioned to be analysed by SEM, while the rest were processed according to the Exakt system and sectioned for light microscopy.

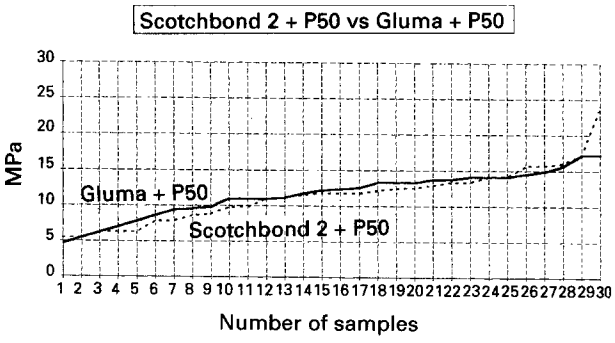

Figure 1 Mean shear bond strength values for Gluma (——) and Scotchbond 2 (– – – –).

TABLE I Mean shear bond strength values for Gluma and Scotchbond 2

Sample number	Scotchbond 2 (MPa)	Gluma (MPa)
1	5.492	4.707
2	5.492	5.492
3	6.276	6.276
4	6.276	7.061
5	6.276	7.845
6	7.845	8.630
7	7.845	9.415
8	8.630	9.611
9	8.826	9.807
10	9.807	10.984
11	9.807	10.984
12	10.984	10.984
13	11.180	11.180
14	11.572	11.768
15	11.768	12.160
16	11.768	12.357
17	11.768	12.553
18	12.160	13.337
19	12.455	13.337
20	12.553	13.337
21	12.945	13.730
22	13.337	13.730
23	13.337	14.122
24	14.122	14.122
25	14.122	14.122
26	15.691	14.514
27	15.691	14.906
28	16.083	15.691
29	17.260	17.260
30	23.536	17.260
Average	11.497	11.709
Standard deviation	3.990	3.263

Student's T-test ($p = 0.915$) showed no significant statistical differences between the two sample groups.

3. Results

The loads required to fracture the specimens at the bone–composite interface were analysed; using a stereomicroscope, adhesive and cohesive fractures were observed. For Gluma the mean of the bone bond strength values was 11.7 MPa (SD 3.2), while for Scotchbond 2 it was 11.5 MPa (SD 3.9) (Fig. 1 and Table I.).

Transmitted normal and polarized light microscopy showed a tight adaptation of the posterior composite to the cortical bone in both specimen groups (Figs 2 and 3).

SEM data confirmed the histological results: no wide gaps or evident composite fractures were present at the bone–composite interface in either specimen group (Figs 4 and 5). All specimens showed a good composite-to-bone adaptation, and no gaps were observed at the bone–composite interface (Figs 6 and 7). In proximity to the bone, only microcracks (1–2 μm) inside the composite were observed (Fig. 8).

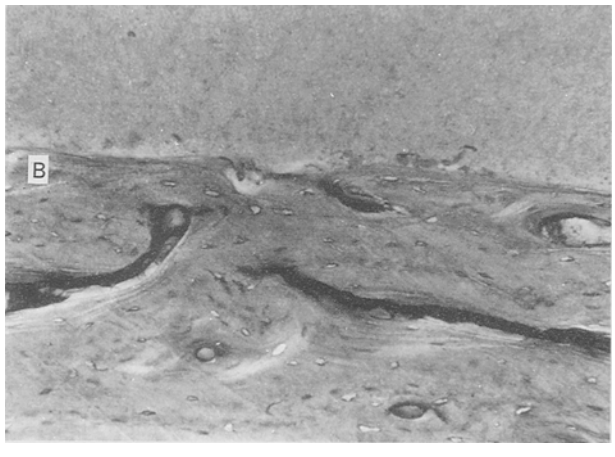

Figure 2 Light microscopy picture of the bone–composite interface using Scotchbond 2 as an adhesive. Good composite adaptation to bone surface (B) is observable (1000 ×).

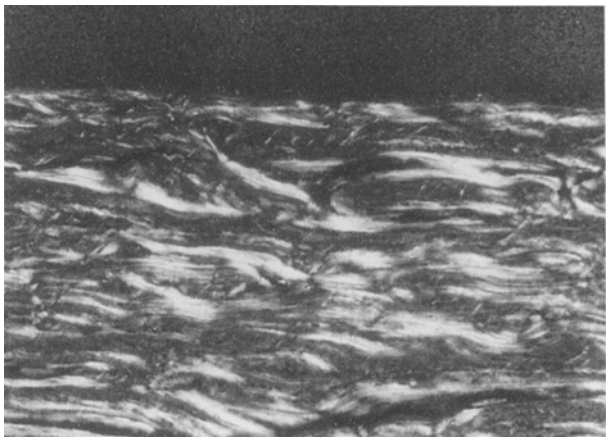

Figure 3 Polarized light microscopy picture of the bone–composite interface using Gluma as adhesive. No evident gap is detectable between composite and bone (1000 ×).

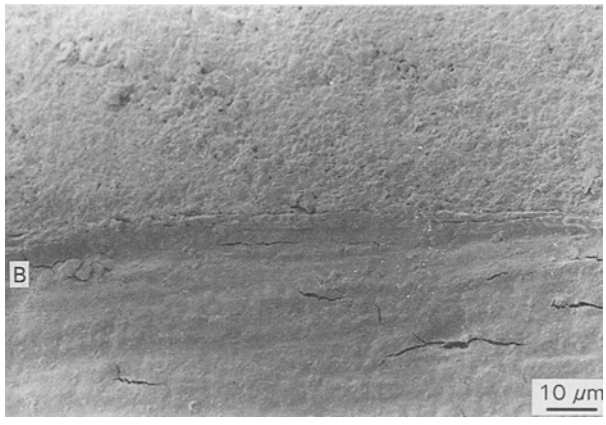

Figure 6 SEM micrograph of the bone–composite interface using Scotchbond 2 as adhesive. Good adaptation of composite to bone and no gaps were detectable at higher magnification.

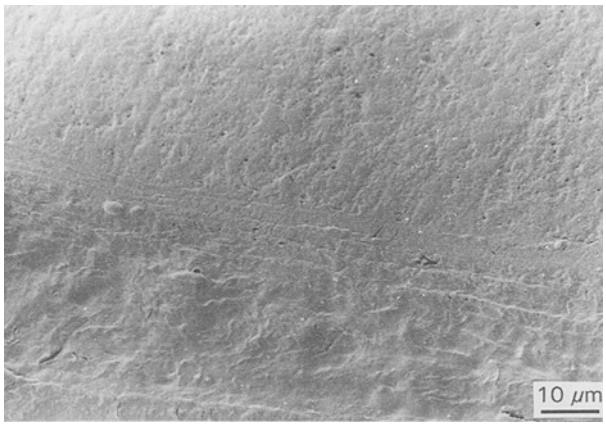

Figure 4 SEM micrograph of the bone–composite interface using Scotchbond 2 as adhesive. No gaps are visible between composite and bone (B).

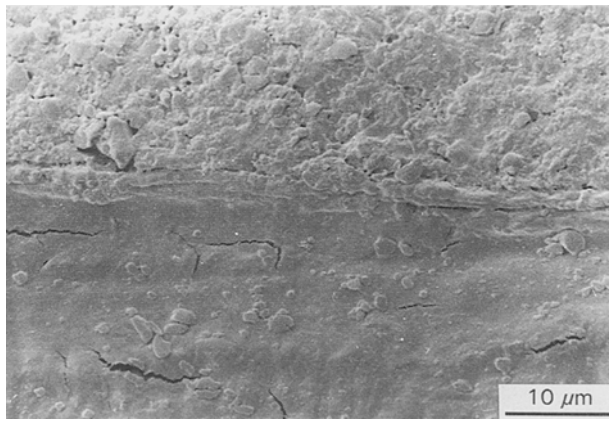

Figure 7 SEM micrograph of the bone–composite interface using Gluma as adhesive. There is no gap between composite and mineralized tissue. On the left an osteocyte is visible in a bone lacuna.

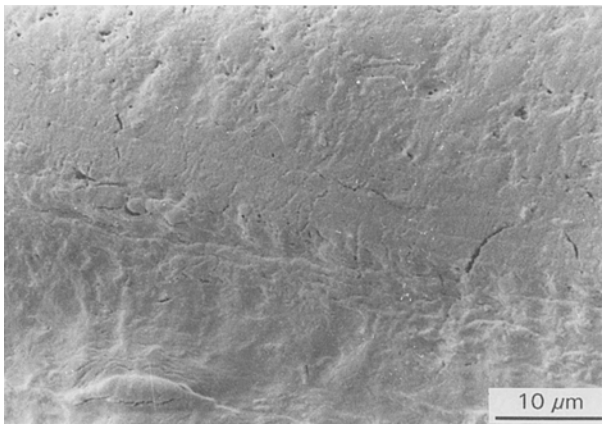

Figure 5 SEM micrograph of the bone–composite interface using Gluma as adhesive. A good adaptation of the posterior composite to the bone surface is observable.

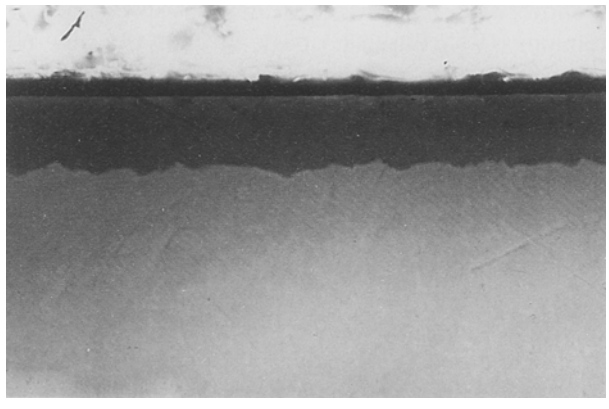

Figure 8 SEM micrograph of the bone–composite interface using Scotchbond 2 as adhesive. In some areas between composite and bone (B) microfractures are observed inside the composite, probably due to the contraction of the composite during polymerization.

In a few areas, gaps of 5–12 μm, often showing cohesive separation, were present in proximity to the bone–composite interface (Fig. 9). Moreover, in the cortical bone it was possible to observe empty

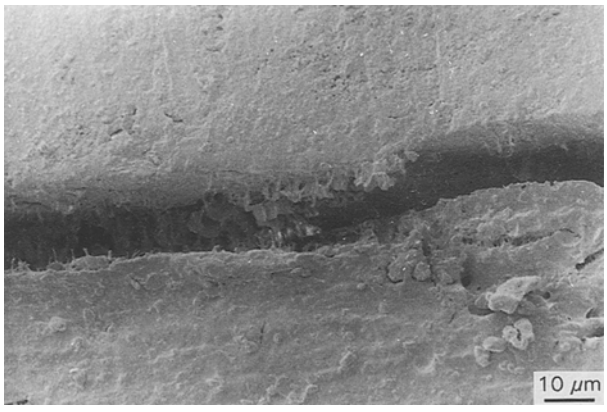

Figure 9 SEM micrograph of the bone–composite interface using Gluma as adhesive. A gap showing cohesive separation, probably due to composite contraction, is appreciable in proximity to the bone–composite interface. Remnants and small particles of the composite bonded to the bone surface suggest cohesive separation.

osteocyte lacunae and Haversian systems filled by the composite in the external surface.

4. Discussion

The analysis of the loads required to fracture the specimens treated with Gluma and Scotchbond 2 shows that the mean values are similar. The two dentin bonding systems, which have different mechanisms [19], allow the formation of a strong bond between a methacrylate-based material and fresh bovine bone tissue [19].

Histological and ultrastructural observations confirm the results of the shear strength test; no wide gaps are observed at the bone–composite interface. Gaps of 5–12 μm and the few microcracks of 1–2 μm inside the composite and in the proximity of the bone surface may be a consequence of composite polymerization contraction [20, 21].

Recently, extending the use of these mathacrylate-based composite materials has been tried, due to their ease of manipulation and their low cost [22]. The use of fresh autogenous grafts in bone reconstruction has limitations and disadvantages, such as limited availability of donor sites, difficulty in accurate shaping and contouring of the graft, resorption of the graft, an additional surgical procedure required to obtain the graft tissue with subsequent donor site morbidity and increased operating time. All these limitations have produced therefore an increased demand for inorganic graft or implant material.

The introduction of inorganic material for surgical applications has brought significant change in the scope and procedures of operations in all surgical specialities [19]. The dental composite materials have been tested by the American Dental Association and are used in everyday dental practice as permanent restorative materials. They are applied directly to dentin and are in direct contact to pulp through the dentinal tubules. They must be considered well tolerated by the body without toxic or other adverse effects.

In this study the shear bond strength test and the ultrastructural observations have demonstrated the strong bonding between a posterior composite and fresh bovine bone using different dentinal bonding adhesives. With Gluma Dentin Bond System and Scotchbond 2 Dental Adhesive System the means of the loads required to fracture the specimens at the bone–composite interface were 11.7 MPa and 11.5 MPa, respectively. Good adaptation of the posterior composite to the cortical bovine femur bone has been observed and no evident gaps at the interface bone–composite were present in all specimens of both groups.

Our results could open the way to the use of methacrylate-based materials, such as dental composites with bonding systems, in an array of surgical fields such as oral and maxillofacial surgery, orthopaedics, ENT, neurosurgery and plastic surgery. Adhesive materials such as glass ionomers have been experimentally used with success in periodontal defects. Clinical and histological studies have demonstrated that these materials are well tolerated by the host tissues and may be considered biocompatible [23].

Moreover bonding between bone and cyanoacrylate polymers, with and without the addition of powdered hydroxyapatite, has been demonstrated *in vitro* [24]. Even if the bonding materials used in this study are probably designed to be used as a thin layer and we have no idea of the mechanical properties of these materials when used as a bulk material for a bone graft, nevertheless the elastic modulus of these composite materials is very similar to human dentin and probably their response to the application of forces is similar to mineralized tissues.

A clinical limitation of the application of posterior composites with a dentinal bonding agent to fresh bone may be bleeding at the bone site. The materials that we have tested are not new and they are commercially distributed as restorative materials for dental practice. We hope to present in the future *in vivo* experiments which may help to clarify if the turnover of the bone can modify the composite bonding over time, and further tests to assess the interactions between the implanted materials and bone.

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