

Fabrication and evaluation of Bis-GMA/TEGDMA resin with various amounts of silane-coated silica for orthodontic use

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SUMMARY The objective of this research was to fabricate a composite with an optimum filler level in a bisphenol-A-glycidyl dimethacrylate (Bis-GMA) triethylene glycidyl dimethacrylate (TEGDMA) resin for bonding of metallic orthodontic brackets to achieve the best handling characteristics with optimum bond strength and without compromising the mechanical properties of the adhesive.

One-hundred and sixty extracted human premolars free of any detectable pathology or buccal surface alterations were collected and divided into four groups. In group 1 (control), the teeth were bonded with stainless steel brackets using Transbond XT. In groups 2, 3, and 4, the teeth were bonded with metal brackets using a Bis-GMA/TEGDMA resin with 80, 60, and 20 per cent by weight silane-coated silica of a spherical shape with a mean size of 0.01 μ m. Shear bond strength (SBS) of the composites was determined and the adhesive remnant index (ARI) and enamel fracture post-debonding were assessed.

According to one-way analysis of variance and Tukey's honestly significant difference (HSD) multiple comparison tests, the SBS of group 4 (10.54 MPa) was considerably less than that of groups 1 (26.1 MPa), 2 (25.5 MPa), and 3 (24.6 MPa). Chi-square analysis revealed that there was an insignificant difference in the incidence of enamel fracture between groups 1 and 2, while a significant difference was present between groups 1 and 2 and 3 and 4. An insignificant difference was also observed in the location of the adhesive failure between the four groups. While all the bonding adhesives tested can be safely used for bonding of brackets, 60 per cent filled Bis-GMA/TEGDMA was superior clinically due to its ease of handling and superior bond strength.

Introduction

The desirable handling characteristics of adhesives such as non-stickiness, fluid injectability, adequate working time, short curing time, high erosion resistance, and a simple bonding procedure with minimal steps involved have not been achieved in a single adhesive. Incorporation of these properties would make an ideal universal composite for direct and indirect bonding of attachments and lingual retainers (Eloughth *et al.*, 2002).

Highly filled orthodontic adhesives with 80 per cent fillers, such as Transbond XT, provide adequate bond strengths but the filler content prevents these adhesives being used for bonding lingual retainers and indirect bonding of attachments, plus the reduced flowability of these adhesives makes it mandatory for the clinician to apply a low-viscosity primer on the tooth surface prior to bonding. Injectable orthodontic adhesive with less than 20 per cent of filler such as Heliosit Orthodontic do not require primer application but have compromised bond strength and poor wear resistance (Stormann and Ehmer, 2002; Murray and Hobson, 2003). Three-step bonding procedures of etching, priming, and composite application are not only time-consuming but there is a high probability of saliva/water contamination. Two-step procedures on the other hand combine etchant and primer in one solution

[self-etching primers (SEPs)] or do not use a primer as in the case of flowable composites (D'Attilio *et al.*, 2005; Tecco *et al.*, 2005). The problem with SEPs is that they are technique sensitive (Bishara *et al.*, 2007), cannot be used with all composites, and have reduced bond strengths and bracket survivability when compared with the phosphoric acid etching technique (Bishara *et al.*, 1999; Aljubouri *et al.*, 2003; Cehreli *et al.*, 2005).

The aim of the present study was to determine the optimum level of filler concentration in a bisphenol-A-glycidyl dimethacrylate (Bis-GMA) triethylene glycidyl dimethacrylate (TEGDMA) resin, by establishing and comparing the bond strengths, adhesive failure location, and enamel fracture post-debonding with that of Transbond XT, with different concentrations of silane-coated silica, yielding composites of different viscosity.

Materials and methods

The experimental adhesives consisted of Bis-GMA/TEGDMA monomer matrix mixed at a ratio of 50:50 (Feilzer and Dauvillier, 2003; Tian *et al.*, 2008), with an initiator (camphorquinone) and functional silane-treated silicone dioxide (SiO₂) of a spherical shape and mean

filler size of 0.01 μ m. Three different adhesives were manufactured keeping the Bis-GMA/TEGDMA ratio constant and varying only the amount of filler. Type 1 composite had a filler content of 80 per cent by weight, type 2 flowable composite with a filler level of 60 per cent by weight, and type 3 a flowable composite consisting of a filler content of 20 per cent by weight. The adhesive matrix was mixed in a speed mixer (Speed Mixer DAC 150FVZ, Hauschild Engineering, Hamm, Germany) for 60 seconds at 1800 rpm to attain an homogenous mixture. The mixtures were degassed in a vacuum chamber prior to storing in opaque containers to avoid premature curing. The adhesives were manufactured 24 hours prior to shear bond testing.

One-hundred and sixty extracted premolar teeth of either arch or side with intact and well-supported enamel, free of any enamel defects, were collected. The extracted teeth were washed and kept in normal saline for a maximum of 1 month to prevent them from drying out. The teeth were randomly distributed into the following four groups of 40 teeth each using the randomization software (Random Allocation Software, Version 1.0.0, Isfahan, Iran). Group 1: teeth bonded with Transbond XT (control), group 2: bonded with type 1 composite, group 3: bonded with type 2 composite, and group 4 bonded with type 3 composite.

A standard procedure was employed for bonding all brackets in group 1. The buccal surface of each tooth was cleaned with a rubber polishing cup and non-fluoridated pumice powder in a slow handpiece (Uysal *et al.*, 2003; Faltermeier *et al.*, 2007a). The tooth was then thoroughly dried. This was followed by etching with 37 per cent phosphoric acid for 15 seconds and rinsing for 30 seconds with water (Carstensen, 1995; Bin Abdullah and Rock, 1996). After air-drying, a thin coat of Heliobond (3M Espe, Monrovia, California, USA) primer was applied with a brush. A stainless steel bracket (Ortho Organizers, San Marcos, California, USA) with Transbond XT on the mesh base was then firmly placed 3.5 mm away from the occlusal surface with a Wicks stick tooth positioning gauge (Falcon Medical Instruments, Sialkot, Pakistan). The force to press the bracket against the tooth was measured with a tension/compression measuring gauge (Dentaurum, Pforzheim, Germany; Arici *et al.*, 2005). This was followed by curing for 20 seconds with a light-emitting-diode-curing unit (Apoza Enterprise Company Limited, Taipei Hsien, Taiwan). The light tip was kept at the buccal cusp tip so that the distance of the light source from the bracket base was the same for each bracket i.e. 3.5 mm. The light intensity of the light curing unit was measured after every 10 cycles at 800 ± 25 mW/cm² using a digital light intensity measuring device (Apoza Enterprise Company Limited). Efforts were made to ensure the intensity of light, time of light curing, the distance of the light source from the bracket base and the thickness of the composite, by keeping the force to place the bracket constant throughout the bonding of the brackets both within and between groups.

For groups 2, 3, and 4, the same protocol was followed as for group 1, except that no primer was applied to the tooth surface prior to the placement of the brackets in groups 3 and 4.

Shear bond strength (SBS) was determined using a universal testing machine (Instron Corporation, Canton, Massachusetts, USA) with a crosshead speed of 0.5 mm/minute and a load range of 0.04–20 kg. The teeth were embedded in circular moulds with a diameter of 26 mm filled with autopolymerizing polymethyl methacrylate (Esschem Co, Portland, Oregon, USA). A mounting jig was used to align the facial surface of the tooth so that it was perpendicular to the base of the mould; this made the labial surface of the tooth parallel to the gingivo-occlusal force when the mould with the tooth was held in the crosshead of the testing machine. A 0.1 inch ligature wire was tied around the wings of the bracket and its free end was engaged in the other crosshead of the testing machine. The load applied at failure was recorded in Newton and converted to stress (force per unit area) in Mega Pascal (MPa). The dimensions of the bracket base were measured with a digital vernier calliper accurate to 0.01 mm (Mititoyu, Miyazakai, Japan).

To determine the adhesive remaining on the tooth surface post-debonding, the adhesive remnant index (ARI; Årtun and Bergland, 1984) was employed. The presence of enamel cracks was assessed by viewing the teeth under $\times 10$ magnification with a stereomicroscope (A.M.D. Dental Mfg., Inc., Nazareth, Pennsylvania, USA).

To control operator bias, the results were verified by two individuals not involved in the study.

Data analysis

The Statistical Package for Social Sciences, version 11.0 (SPSS Inc. Chicago, Illinois, USA), was used for statistical analysis. The variables in this study were shear force per unit area measured in MPa, ARI score, and the presence/absence of enamel fracture post-debonding. Descriptive statistics, including the mean, standard deviation, range, variance, minimum and maximum of SBS (MPa), as well as frequency distribution of the ARI scores (percentage) and presence of sound/fractured enamel (percentage) were calculated. One-way analysis of variance (ANOVA) and Tukey's honestly significant difference (HSD) multiple comparison tests were used to determine the statistical difference between the independent variable (SBS) and the four dependent variables (groups 1, 2, 3, and 4). A chi-square test was used to evaluate differences in the frequency distribution of ARI score and enamel fracture. Significance for all tests was predetermined at $P \leq 0.05$.

Results

Shear bond strength

One-way ANOVA indicated that the mean SBS of groups 1, 2, 3, and 4 was 26.1 ± 1.56 , 25.5 ± 1.69 , 24.6 ± 2.58 , and

10.54 \pm 1.87 MPa, respectively (Figure 1a). The *post hoc* HSD test for multiple comparisons revealed that there was an insignificant difference between the SBS of groups 1, 2, and 3, while there was a significant difference between the bond strengths of group 4 and groups 1, 2, and 3.

Enamel fracture

Thirteen teeth in group 1 fractured at the enamel interface during debonding, which accounted for 32.5 per cent of the bonded teeth in that group, while in 12 teeth in group 2, enamel fracture was detected (30 per cent). The enamel fractured in seven teeth in group 3 (17.5 per cent) and in four teeth in group 4 (10 per cent; Figure 1b). There was an insignificant difference in the incidence of enamel fracture between groups 1 and 2, while a significant difference was present between groups 1 and 2 and groups 3 and 4.

Adhesive remnant index

The results of the ARI are summarized in Figure 1c and 1d. There was an insignificant difference between the ARI

scores of the four groups. The maximum number of teeth in all the groups scored 3 i.e. adhesive failures occurred at the bracket-adhesive interface.

Discussion

The physical, mechanical, and aesthetic properties and clinical behaviour of composites depend on their structure. Dental composites are composed of four chemically different materials: organic matrix or organic phase, inorganic matrix i.e. filler or dispersal phase, the initiator accelerator system, and the organosilane or coupling agent to bond the filler to the organic resin (Goldstein, 2002).

Bis-GMA is the most commonly used monomer in contemporary composites; it has a very large molecular weight and is highly viscous which gives the composite stability but the viscosity makes the manufacturing processes and clinical handling more difficult. To facilitate manufacturing, it is mixed with low-viscosity monomers such as TEGDMA, ethylene glycol dimethacrylate, methyl methacrylate, or urethane dimethacrylate (Culbertson *et al.*, 1997; Holter *et al.*, 1997).

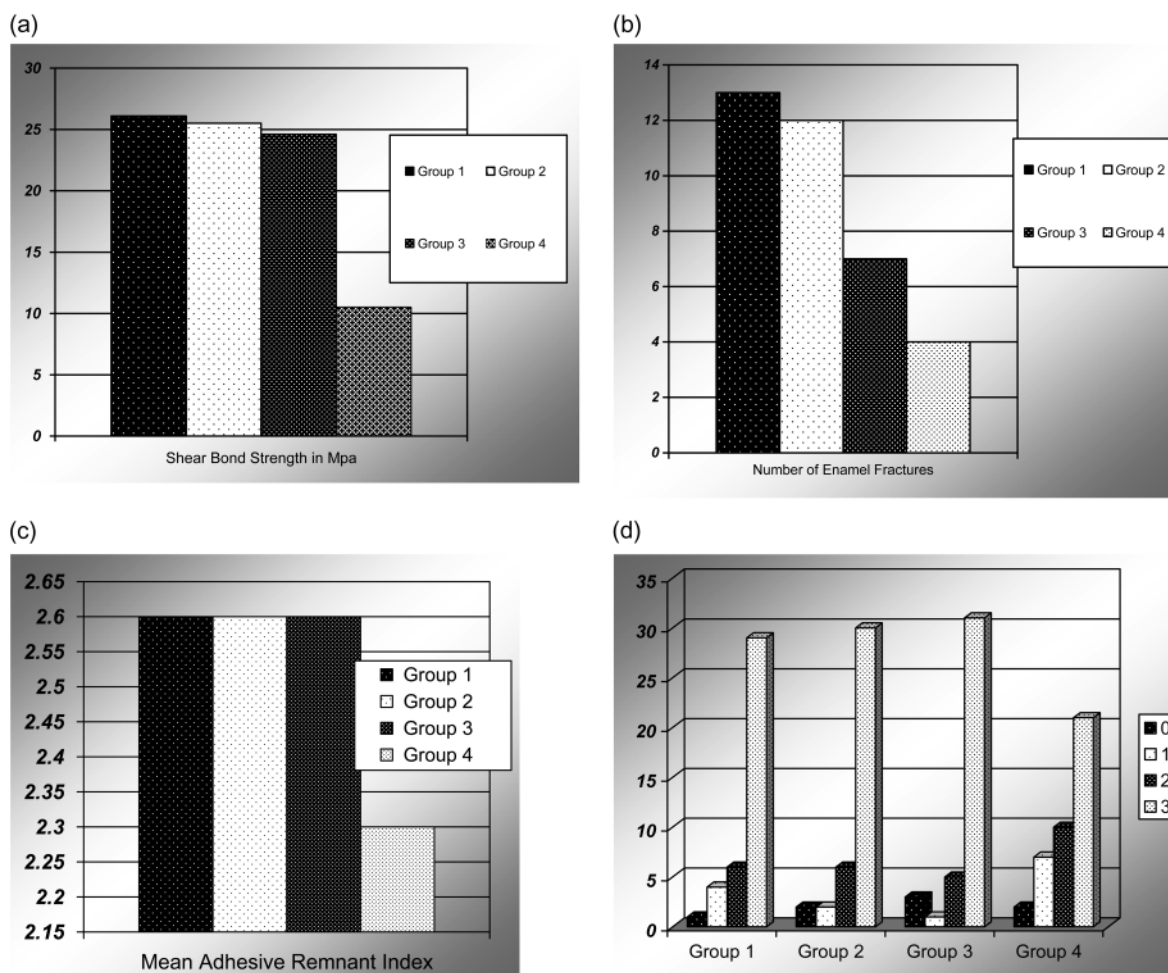


Figure 1 Graphs showing (a) the mean shear bond strength of the composites used. (b) The number of teeth in the different groups with enamel fractures post-debonding. (c) The mean adhesive remnant index (ARI) scores for the different groups. (d) The ARI scores for individual teeth in the different groups.

The dispersal phase of composite resins is made up of an inorganic filler material that, in essence, determines the physical and mechanical properties of the composite. The filler reduces the coefficient of thermal expansion and overall curing shrinkage, provides radio-opacity, improves handling and cohesion within the material, and improves the aesthetic result (Labella *et al.*, 1999). The filler particles used vary widely in their chemical composition, morphology, and dimensions. The various fillers employed are boron silicates and lithium aluminium silicates.

Flowable composites are low-viscosity resins, making them more fluid than conventional composite resins and because of a lower filler load they are also known as 'wettable composites'. A flowable composite typically contains 42–62 per cent of inorganic fillers in contrast to packable composites, which may have a filler concentration as high as 80 per cent (Powers and Sakaguchi, 2007).

The main advantages of flowable composites in orthodontics are high wettability of the tooth and the bracket surface, ensuring penetration into irregularities and elimination of the need of a bonding agent on the tooth surface prior to bonding, and its ability to form layers of minimum thickness, improving or eliminating air inclusion or entrapment. Moreover, flowable composites have high flexibility making them less likely to be displaced in stress concentration areas, injectability eases the application of the composite, and flowability allows for the indirect bonding of the bracket with a minimum formation of flash (Olmez *et al.*, 2004).

There have been a limited number of studies on the use of flowable composites to bond brackets. Uysal *et al.* (2004), after comparing the SBS of three commercially available flowable composite, criticized the use of flowable composites to bond orthodontic brackets because of their inadequate bond strengths.

The aim of this research was to fabricate a composite that was sufficiently flowable to be used in an injectable form without the use of an intermediary resin and without compromising bond strength and the safety of the adhesive. Most bond strength studies use commercially available adhesive systems that have different filler particle sizes and concentrations with various resins. This makes comparisons difficult because of the considerable number of variables involved in the adhesive composition (Ostertag *et al.*, 1991). All the variables in the present study were kept constant, except for the level of filler, to ascertain the effect of filler concentration on the adhesive compared with a control (Transbond XT). Intensity of light (Signorelli *et al.*, 2006; Staudt *et al.*, 2006), length of light curing (Silta *et al.*, 2005; Gronberg *et al.*, 2006), and the distance of light (Cacciafesta *et al.*, 2005; Gronberg *et al.*, 2006; Sfondrini *et al.*, 2006) source from the bracket base and the thickness of the composite (by keeping the force to place the bracket uniform) were also kept constant throughout bonding of the brackets both within and between groups as these can have an effect on the SBS of the composites.

The filler content of commonly used orthodontic adhesives; Transbond XT (80 per cent by weight) and Heliosit Orthodontic (14–20 per cent by weight) were used as a guide in the fabrication of types 1 and 3 adhesives in the current study. Type 2 adhesive was manufactured keeping in mind that increasing the filler level increases the bond strength of metal brackets to the tooth at the same time as decreasing the flowability of the material (Ostertag *et al.*, 1991; Faltermeier *et al.*, 2007b). A pilot study was carried out prior to this experiment where composites with 20, 40, 60, and 80 per cent were tested on 15 teeth each. These results showed that a filler content of 60 per cent by weight would be ideal to achieve adequate flowability and strength. It was also desired that the two flowable composites, types 2 and 3, could be used without an intermediary unfilled resin to ease the bonding procedure and reduce the risk of saliva contamination (Uysal *et al.*, 2004; Tecco *et al.*, 2005).

The SBS increased considerably with an increase in the filler content from 20 (10.54 MPa) to 60 (24.6 MPa) per cent, in agreement with the findings of Faltermeier *et al.* (2007b) and Ostertag *et al.* (1991). An insignificant difference between the SBS of groups 3 and 2 (25.5 MPa), and 1 (26.1 MPa) was observed, indicating that a flowable composite can be used for orthodontic bonding of metal brackets without compromising SBS.

For most of the brackets in groups 1 (75 per cent), 2 (75 per cent), and 3 (76 per cent), adhesive failure at the bracket-adhesive interface occurred, indicated by the ARI score of 3. Fifty-three per cent of the brackets in group 4 showed failure at the bracket-adhesive interface. These results may be due to the higher filler content in groups 1, 2, and 3 which increases cohesion within the cement. Furthermore, the lower viscosity of the composite in group 4 enabled it to more efficiently wet the undercuts under the bracket and tooth, thus increasing the adhesion of the composite to the bracket base (Faltermeier *et al.*, 2007b).

There was a relationship between SBS and the frequency of enamel fracture. Group 1 had an SBS of 26.1 MPa and the enamel fractured in 32.5 per cent of teeth, and in group 2 with an SBS of 25.52 MPa, the enamel fractured in 30 per cent. This percentage decreased to 17.5 for group 3 (SBS = 24.59 MPa) and 10 for group 4 (SBS = 10.51 MPa). There was a clear relationship between SBS and the presence of enamel fracture. The greater the bond strength the more stress is generated on and within the tooth surface during debonding of the bracket, which in turn leads to enamel fracture. This is why highest fracture rates were observed in groups 1 and 2 and the lowest in group 4 (Figure 1c and 1d).

The results of the present study showed that Bis-GMA/TEGDMA composite with 60 per cent filler can be used for bonding orthodontic brackets without an intermediary low-viscosity resin while reducing the working time and the possibility of fluid contamination when compared with conventional Transbond XT. Additionally, the flowability of such a composite makes it ideal for indirect bonding at the

same time making the clinical procedure easier for the bonding of lingual retainers. The results for the composite with a filler content of 20 per cent were not as promising; even though it demonstrated a lower bond strength than the other two groups, it can be used for clinical use as the mean SBS was higher than the 6–8 MPa recommended values of Reynolds (1975, 1976).

Conclusions

1. The SBS of all composites tested were greater than 6–8 MPa, with the flowable composites showing a significant difference in the frequency of enamel fractures.
2. There was an insignificant difference between the SBS of groups 1, 2, and 3, while a significant difference in the SBS of groups 1, 2, 3, and 4 was present. Group 4 showed the lowest SBS
3. In groups 1, 2, and 3, the most common site of composite fracture was at the bracket composite interface.
4. Bis-GMA/TEGDMA with 60 per cent silane-coated spherical SiO₂ with a mean diameter of 0.01 μ m showed promising results. It has the advantages of flowable composites and the bond strength of highly filled packable composites.

References

- Aljubouri Y D, Millet D T, Gilmour W H 2003 Laboratory evaluation of a self-etching primer for orthodontic bonding. *European Journal of Orthodontics* 25: 411–415
- Arici S, Caniklioglu C M, Arici N, Ozer M, Oguz B 2005 Adhesive thickness effects on the bond strength of a light-cured resin-modified glass ionomer cement. *Angle Orthodontist* 75: 254–259
- Årtun J, Bergland S 1984 Clinical trials with crystal growth conditioning as an alternative to acid etch enamel pretreatment. *American Journal of Orthodontics* 85: 333–340
- Bin Abdullah M, Rock W P 1996 The effect of etch time and debond interval upon the shear bond strength of metallic orthodontic brackets. *British Journal of Orthodontics* 23: 121–124
- Bishara S E, Gordan V V, VonWald L, Jakobson J R 1999 Shear bond strength of composite, glass ionomer, and acidic primer adhesive systems. *American Journal of Orthodontics and Dentofacial Orthopedics* 115: 24–28
- Bishara S E, Ostby A W, Laffoon J F, Warren J J 2007 The effect of modifying the self-etchant bonding protocol on the shear bond strength of orthodontic brackets. *Angle Orthodontist* 77: 504–508
- Cacciafesta V, Sfondrini M F, Scribante A, Boehme A, Jost-Brinkmann P G 2005 Effect of light-tip distance on the shear bond strengths of composite resin. *Angle Orthodontist* 75: 386–391
- Carstensen W 1995 Effect of reduction of phosphoric acid concentration on the shear bond strength of brackets. *American Journal of Orthodontics and Dentofacial Orthopedics* 108: 274–277
- Cehreli Z C, Kecik D, Kocadereli I 2005 Effect of self-etching primer and adhesive formulations on the shear bond strength of orthodontic brackets. *American Journal of Orthodontics and Dentofacial Orthopedics* 127: 573–579
- Culbertson B, Wan Q, Tong Y 1997 Preparation and evaluation of visible light-cured multi-methacrylates for dental composites. *Journal of Macromolecular Science: Pure and Applied Chemistry* 34: 2405–2421
- D'Attilio M, Traini T, Di Iorio D, Varvara G, Festa F, Tecco S 2005 Shear bond strength, bond failure, and scanning electron microscopy analysis of a new flowable composite for orthodontic use. *Angle Orthodontist* 75: 410–415
- Elaugh J, Asscherickx K, Vande Vannet B, Wehrbein H 2002 Flowable composites for bonding lingual retainers. *Journal of Clinical Orthodontics* 36: 597–598
- Faltermeier A, Behr M, Müssig D 2007a A comparative evaluation of bracket bonding with 1-, 2-, and 3-component adhesive systems. *American Journal of Orthodontics and Dentofacial Orthopedics* 132: 144.e1–e5
- Faltermeier A, Rosentritt M, Faltermeier R, Reicheneder C, Müssig D 2007b Influence of filler level on the bond strength of orthodontic adhesives. *Angle Orthodontist* 77: 494–498
- Feilzer A J, Dauvillier B S 2003 Effect of TEGDMA/BisGMA ratio on stress development and viscoelastic properties of experimental two-paste composites. *Journal of Dental Research* 82: 824–828
- Goldstein R 2002 *Sistemas adhesivos de los composites*. STM Editores, Barcelona
- Gronberg K, Rossouw P E, Miller B H, Buschang P 2006 Distance and time effect on shear bond strength of brackets cured with a second-generation light-emitting diode unit. *Angle Orthodontist* 76: 682–688
- Holter D, Frey H, Mulhaupt R 1997 Branched bismethacrylates based on Bis-GMA—a systematic route to low shrinkage composites. *Polymer Preprints* 38: 84–85
- Labella R, Lambrechts P, Van Meerbeek B, Vanherle G 1999 Polymerization shrinkage and elasticity of flowable composites and filled adhesives. *Journal of Dental Materials* 15: 128–137
- Murray S D, Hobson R S 2003 Comparison of *in vivo* and *in vitro* shear bond strength. *American Journal of Orthodontics and Dentofacial Orthopedics* 123: 2–9
- Olmez A, Oztas N, Bodur H 2004 The effect of flowable resin composite on microleakage and internal voids in Class II composite restorations. *Journal of Operative Dentistry* 29: 713–719
- Ostertag A J, Dhuru V B, Ferguson D J, Meyer Jr R A 1991 Shear, torsional, and tensile bond strengths of ceramic brackets using three adhesive filler concentrations. *American Journal of Orthodontics and Dentofacial Orthopedics* 100: 251–258
- Powers J M, Sakaguchi R L 2007 *Craig's restorative dental materials*. Mosby, New Delhi
- Reynolds I R 1975 A review of direct orthodontic bonding. *British Journal of Orthodontics* 2: 171–178
- Reynolds I R 1976 A review of direct orthodontic bonding. *British Journal of Orthodontics* 3: 91–95
- Sfondrini M F, Cacciafesta V, Scribante A, Boehme A, Jost-Brinkmann P G 2006 Effect of light-tip distance on the shear bond strengths of resin-modified glass ionomer cured with high-intensity halogen, light-emitting diode, and plasma arc lights. *American Journal of Orthodontics and Dentofacial Orthopedics* 129: 541–546
- Signorelli M D, Kao E, Ngan P W, Gladwin M A 2006 Comparison of bond strength between orthodontic brackets bonded with halogen and plasma arc curing lights: an *in-vitro* and *in-vivo* study. *American Journal of Orthodontics and Dentofacial Orthopedics* 129: 277–282
- Silta Y T, Dunn W J, Peters C B 2005 Effect of shorter polymerization times when using the latest generation of light-emitting diodes. *American Journal of Orthodontics and Dentofacial Orthopedics* 129: 744–748
- Staudt C B, Krejci I, Mavropoulos A 2006 Bracket bond strength dependence on light power density. *Journal of Dentistry* 34: 498–502
- Stormann I, Ehmer U 2002 A prospective randomized study of different retainer types. *Journal of Orofacial Orthopedics* 63: 42–50
- Tecco S, Traini T, Caputi S, Festa F, de Luca V, D'Attilio M 2005 A new one-step dental flowable composite for orthodontic use: an *in vitro* bond strength study. *Angle Orthodontist* 75: 672–677
- Tian M, Gao Y, Liu Y, Liao Y, Hedin N E, Fong H 2008 Fabrication and evaluation of Bis-GMA/TEGDMA dental resins/composites containing nano fibrillar silicate. *Journal of Dental Materials* 24: 235–243
- Uysal T, Basciftci F A, Uşümez S, Sari Z, Buyukerkmen A 2003 Can previously bleached teeth be bonded safely? *American Journal of Orthodontics and Dentofacial Orthopedics* 123: 628–632
- Uysal T, Sari Z, Demir A 2004 Are the flowable composites suitable for orthodontic bracket bonding? *Angle Orthodontist* 74: 697–702

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