©Operative Dentistry, 2009, **34-6**, 697-702

Properties of Hybrid Resin Composite Systems Containing Prepolymerized Filler Particles

JT Blackham • KS Vandewalle • W Lien

Clinical Relevance

The incorporation of prepolymerized-filler particles in hybrid resin composite systems may result in a reduction of mechanical properties.

SUMMARY

This study compared the properties of newer hybrid resin composites with prepolymerized-filler particles to traditional hybrids and a microfill composite. The following properties were examined per composite: diametral tensile strength, flexural strength/modulus, Knoop microhardness and polymerization shrinkage. Physical properties were determined for each composite group (n=8), showing significant differences between groups per property (*p*<0.001). In general, the traditional hybrid composites (Z250, Esthet-X) had higher strength, composites containing pre-polymerized fillers (Gradia Direct Posterior, Premise) performed more moderately and the microfill composite (Durafill VS) had lower strength. Premise and Durafill VS had the lowest polymerization shrinkage.

Jason T Blackham, DMD, USAF, General Dentistry, Tyndall AFB, Panama City, FL, USA

*Kraig S Vandewalle, DDS, MS, USAF, director, Dental Research, Lackland AFB, San Antonio, TX, USA

Wen Lien, DMD, MS, USAF, General Dentistry, Lackland AFB, San Antonio, TX, USA

*Reprint request: 1615 Truemper St, Lackland AFB, San Antonio, TX 78236, USA; e-mail: kraig.vandewalle@lackland.af.mil

DOI: 10.2341/08-118-L

INTRODUCTION

The recent proliferation of composite materials on the market today may lead to confusion and indecision when selecting a system. However, in spite of the vast number of dental composite products that have appeared over the past 20 years, all of them have simple, common characteristics. Dental resin composites primarily contain a mixture of soft, organic resin matrix (polymer) and hard, inorganic filler particles (ceramic). Filler particles drastically improve the mechanical properties of the composite material. Various classification systems for resin composites have been developed over the years, based on filler particle size.[1] Microfills were developed to provide improved esthetics and polishability.[2] However, in order to maximize filler loading and minimize viscosity, the use of prepolymerized resin and microfiller is used. Mechanical properties, such as strength and stiffness, are generally inferior with microfill composites, because the overall filler content remains low despite the use of pre-polymerized particles.[2]

The most common filler today is barium silicate glass, with an average particle size of 0.6 to 1.0 microns.[1] A small amount of microfiller is added to improve handling characteristics and reduce stickiness.[2] To incorporate a maximum amount of filler into a resin matrix, a distribution of particle sizes is necessary. These so-called hybrids are potentially superior, because increased filler loading improves the stress transfer between particles in the composite.[1] The goal of the manufacturers is to maximize filler loading and minimize filler size, as found with hybrids.[2] Recent developments in trimodal technologies and pre-polymerized materials have resulted in the marketing of newer hybrid resin composite materials. The unique characteristics of these newer hybrid systems are the pre-polymerized particles that are made up of a blend of resin and filler particles.[1]

Recently, a trimodal resin composite (Premise, SDS/Kerr Corporation, Orange, CA, USA) has been marketed. The resin matrix is made up of ethoxylated Bis-GMA and TEGDMA 84% filled by weight with three different filler types: non-agglomerated silica nanoparticles (0.02 µm), pre-polymerized filler (30 µm to 50 µm) and barium silicate glass (0.4 µm). The resin matrix also has light-cure initiators and stabilizers and is claimed to have ultra-low shrinkage with high filler load, sustained polish, excellent handling, non-sticky manipulation and universal application.[3]

Additionally, a microfilled hybrid restorative composite (Gradia Direct Posterior, GC America, Inc, Alsip, IL, USA) has been recently marketed. It contains UEDMA resin matrix 77% filled by weight with fluoro-alumino-silicate glass particles and silica dioxide pre-polymerized filler particles (average 16 µm), with an overall average particle size of 0.85 µm. Gradia Direct Posterior (GC America, Inc) reportedly provides outstanding esthetics without sacrificing strength, durability and longevity.[4]

The objective of the current study was to compare the properties of several representative hybrid and microfill resin composite restorative materials to the new hybrid systems containing pre-polymerized particles. A study by Mitra and others compared the mechanical properties of several resin composite restorative materials and found that, of the materials tested, the hybrid composites were significantly stronger than a microfill composite.[5] No studies to date have been published that evaluate the properties of new hybrid resin composite restorative materials containing pre-polymerized filler particles. The null hypothesis to be tested is that the newer hybrid resin composite restorative materials containing pre-polymerized filler particles have similar properties compared to contemporary hybrid and microfilled systems.

METHODS AND MATERIALS

The following resin composite restorative groups were evaluated—two hybrids (Esthet-X, Dentsply, Milford, DE, USA; and Filtek Z250, 3M ESPE, St Paul, MN, USA), one microfill (Durafill VS, Kulzer, Armonk, NY, USA) and two new hybrid systems containing pre-polymerized particles (Premise, SDS/Kerr, Orange, CA, USA; Gradia Direct Posterior, GC America, Alsip, IL, USA) (Table 1). The following properties were exam-

Table 1: *Listing of Composite Brand, Type, Manufacturer and Basic Filler and Resin Components*

Composite	Type	Manufacturer	Resin	Filler	Wt%	Vol%	Size
Z250	Universal Hybrid	3M ESPE, St Paul, MN, USA	Bis-GMA, UDMA, TEGDMA, Bis-EMA	Zirconia, silica	78	60	0.01-3.5 microns
Esthet-X	Micro Hybrid	Dentsply, Milford, DE, USA	Bis-GMA, Bis-EMA, TEGDMA	Barium silicate glass, silica dioxide	77	60	0.02-2.5 microns
Premise	Nanofilled Hybrid	SDS/Kerr, Orange, CA, USA	Bis-GMA, TEGDMA	Barium silicate glass, silica, prepolymerized filler	84	71	0.02-50 microns
Gradia Direct (Posterior)	Microfilled Hybrid	GC America, Alsip, IL, USA	UEDMA	Barium silicate glass, silica dioxide, prepolymerized filler	77	65	avg: 0.85 microns
Durafill VS	Microfill	Kulzer, Armonk, NY, USA	UEDMA	Silicon dioxide, prepolymerized filler	50.5	40	0.02-0.07 microns

ined per composite: diametral tensile strength, flexural strength/modulus, Knoop microhardness and polymerization shrinkage. Eight specimens were prepared for each of the five restorative materials for each property tested (n=8).

For diametral tensile strength (DTS) testing, a stainless-steel mold (Sabri, Downers Grove, IL, USA) 3-mm in length and 4-mm in diameter, was placed on a Mylar-strip covered glass slide. The restorative materials were packed into the mold and completely filled. The top circular surface of the mold was then covered with a Mylar strip and glass slide. Both the top and bottom of the specimen were exposed to a visible-light polymerization unit (Bluephase 16i, Ivoclar, Amherst, NY, USA) for 20 seconds at high irradiance (1600 mW/cm^2). The specimen was removed from the mold and light cured on each longitudinal side of the cylindrical specimen for 20 seconds—for a total of 80 seconds for each specimen. The specimens were stored in 37°C distilled water for 24 hours. At 24 hours, each specimen was tested by placing the specimen on its longitudinal side on the platens of a universal testing machine (Model 5543, Instron, Canton, MA, USA). The specimens were loaded to failure in compression at a crosshead speed of 1.0 mm/minute. The diametral tensile strength was calculated based on peak load, diameter and length of the specimen.

To determine flexural strength and modulus, the restorative material was placed in a 2 mm x 2 mm x 25 mm stainless-steel mold (Sabri) on a Mylar-strip covered glass slide according to ISO 4049.[6] The surface of the mold was covered as before and exposed to a light polymerization unit for 20 seconds in five separate overlapping increments on both sides. The specimens were stored as before and tested on a three-point bending device using a universal testing machine as before at a crosshead speed of 0.75 mm/minute. The flexural strength was calculated based on peak load, length and area of the specimen. The flexural modulus was determined from the slope of the linear region of the load-deflection curve.

For surface microhardness testing, a polytetrafluoroethylene mold (Sabri) 2 mm in width and 8 mm in diameter was used to prepare each specimen as before. The restorative materials were packed in the mold and the top and bottom surfaces were exposed to the light polymerization unit for 20 seconds as before. The specimens were stored as before until testing using a diamond indenter (LM 300AT, Leco, St Joseph, MI, USA) with a 200-gram load and a 10-second dwell time. Three hardness measurements were made for each specimen and they were averaged.

To determine polymerization shrinkage, the specimens were shaped into a semi-sphere and placed on the polytetrafluoroethylene pedestal in the light path in the

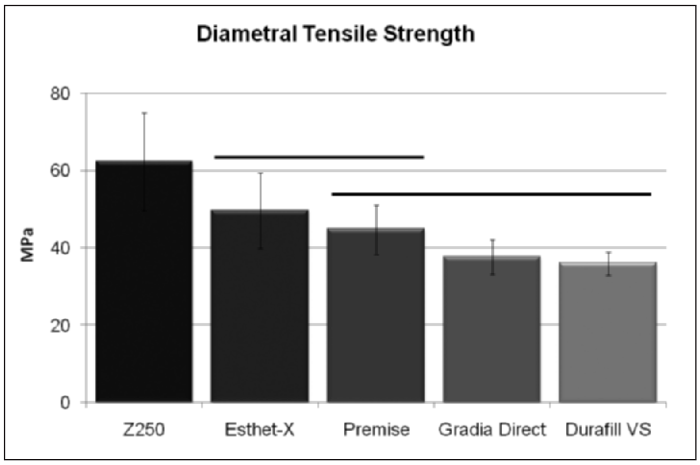

Figure 1. *Bar graph illustrating mean Diametral Tensile Strength (MPa) and standard deviation. Groups joined by the horizontal line are not significantly different (*p*>0.05).*

video-imaging device (AcuVol, BISCO, Schaumberg, IL, USA). The specimens were digitized with a video camera and analyzed with the provided image-processing software. The specimens were allowed to rest for three minutes before being light cured for 20 seconds using the light polymerization unit as before. Polymerization shrinkage was recorded for five minutes after the cure.

The mean and standard deviation were calculated for each of the five mechanical property tests for each of the five restorative materials. A one-way ANOVA followed by the Tukey's post-hoc pairwise comparison test was used to determine whether significant differences existed between the groups per test type at the 0.05 level of significance using statistical software (SPSS, version 10, SPSS Inc, Chicago, IL, USA). For one-way ANOVA with five groups and eight specimens per group, the power was 80% for detecting a moderate effect size (0.58 or 1.16 st dev difference) among means at the alpha level of 0.05.

RESULTS

The one-way ANOVA found significant differences between groups per property (p<0.001). Figure 1 displays the results for diametral tensile strength. Filtek Z250 had the highest DTS. There was no significant difference between Esthet-X and Premise with moderate strength values (p=0.746). Also, there was no significant difference between Premise, Gradia Direct Posterior and Durafill VS at the lower strength values (p=0.219). Figure 2 displays the results for flexural strength testing. Filtek Z250 had the highest strength. Hybrid composites containing pre-polymerized filler particles (Premise, Gradia Direct Posterior) performed more moderately and were not significantly different (p=1.0). The microfill composite (Durafill VS) had the lowest strength. Similar results were found for the flexural modulus (Figure 3). No difference was found

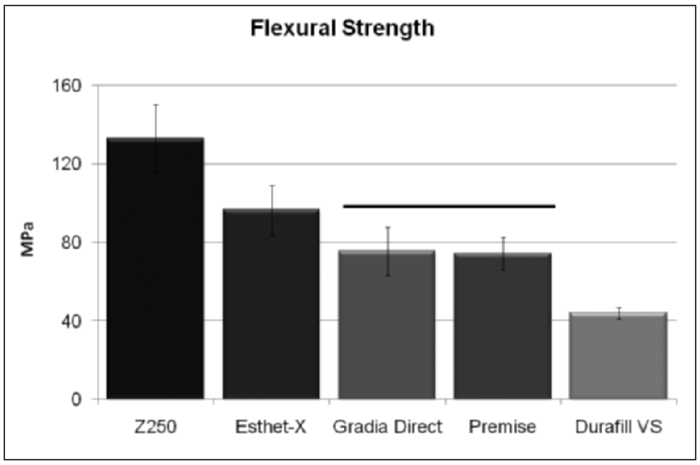

Figure 2. *Bar graph illustrating mean Flexural Strength (MPa) and standard deviation. Groups joined by the horizontal line are not significantly different (*p*>0.05).*

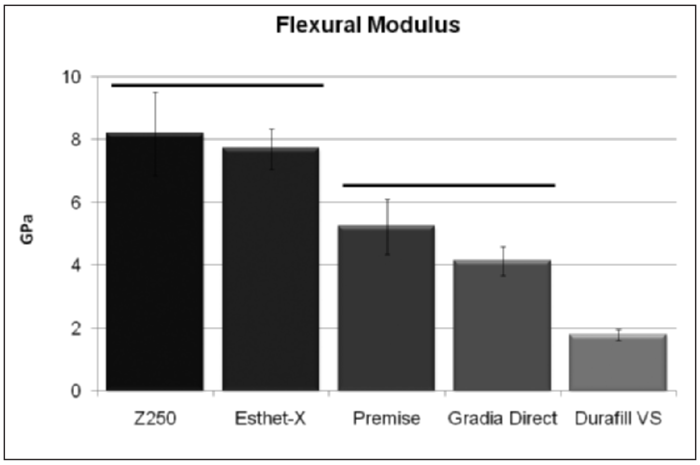

Figure 3. *Bar graph illustrating mean Flexural Modulus (GPa) and standard deviation. Groups joined by the horizontal line are not significantly different (*p*>0.05).*

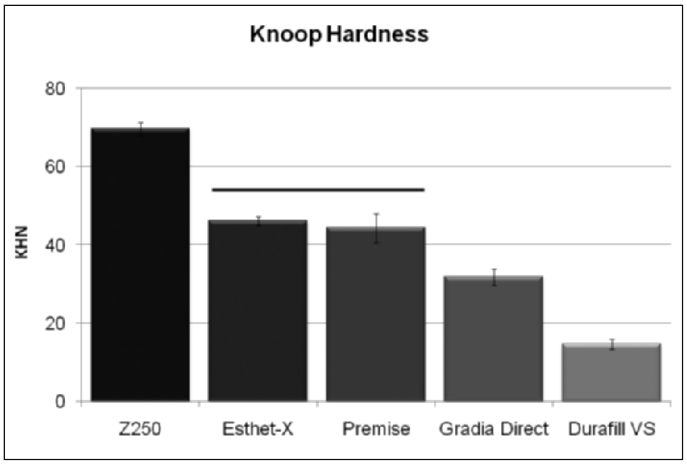

Figure 4. *Bar graph illustrating mean Knoop Hardness Number (KHN) and standard deviation. Groups joined by the horizontal line are not significantly different (*p*>0.05).*

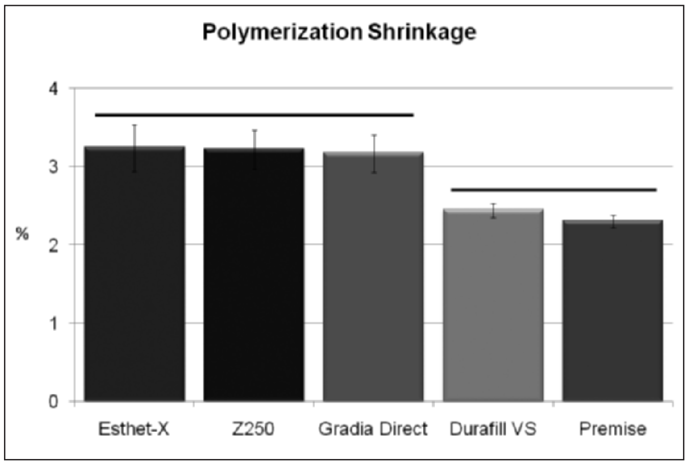

Figure 5. *Bar graph illustrating mean Polymerization Shrinkage (%) and standard deviation. Groups joined by the horizontal line are not significantly different (*p*>0.05).*

between Filtek Z250 and Esthet-X (p=0.740) and between Premise and Gradia Direct Posterior (p=0.071). Figure 4 displays the results for Knoop microhardness. Filtek Z250 had the highest surface hardness. There was no significant difference between Esthet-X and Premise with moderate hardness values (p=0.451). Gradia Direct Posterior had a lower hardness value, with Durafill VS having the lowest hardness value. Figure 5 displays the results for polymerization shrinkage. No difference was found among Esthet-X, Filtek Z250 and Gradia Direct Posterior (p=0.960) and between Durafill VS and Premise (p=0.675).

DISCUSSION

Five commercially available resin composite restorative materials were evaluated in the current study. A significant variation in size, loading and constitution of the filler particles can be seen in the different commercial resin composites tested. The null hypothesis was rejected. Depending on the test, the newer hybrid resin composite restorative materials containing prepolymerized filler particles had different properties compared to the traditional hybrid and microfill systems.

Direct restorative resin composites undergo considerable mechanical challenge during function, especially posterior restorations.[7] In order to withstand higher mechanical stresses, resin composites contain a high percentage of inorganic filler particles.[8-9] Properties, such as strength, hardness and modulus, improve as filler content increases. However, in some cases, an optimum value may be reached at a certain filler volume-fraction, then stabilizes or declines with any further increase.[10]

A resin composite with a high modulus, along with high fracture properties, such as flexural strength, diametral tensile strength and fracture toughness, may be able to better resist occlusal loads.[11] As expected, the highly filled hybrid composite Filtek Z250 had the highest diametral tensile and flexural strength and microhardness of the materials tested due to its wide range of filler particles that include large particles (3.5 microns). The larger filler particles may strengthen the resin composite.[12] Filler size is only one of several factors affecting the properties of resin composites. The type, shape, amount and coupling to resin matrix of filler particles may also affect performance. Smooth, spherical-shaped filler particles are related to an increased packing of the particles and they contribute to the higher strength of Z250, which contains round-shaped particles.[8,13] In addition, the differences in strength among the four hybrid resin composites tested may be due to other factors, since the filler loading was similar (see Table 1). Another factor, in addition to filler content, is stress transfer from the resin matrix to filler particles. The use of pre-polymerized filler particles with Premise, Gradia Direct Posterior and Durafil VS could have contributed to the lower flexural strength and modulus.[1]

Hardness, a measure of the resistance to plastic deformation, is indicative of the ease of finishing of the restoration and its resistance to scratching. Scratches can compromise fatigue strength and lead to premature failure.[14] The Knoop hardness of hybrid resin composites is typically greater than that of microfill resin composites, because of the hardness and volume fraction of the individual filler particles. Knoop hardness values indicate a moderate resistance to indentation under functional stresses for more highly filled composites.[14] In the current study, as expected, the microfill composite Durafil VS had the lowest surface hardness. Gradia Direct Posterior, which also contains pre-polymerized filler particles, had a relatively low hardness value.

AcuVol is a relatively new method of measuring the volumetric polymerization shrinkage of resins using video imaging. This technique has been shown to yield results comparable to those observed using mercury dilatometry[15] and drop-shape analysis.[16] Polymerization shrinkage has been an inherent deficiency with resin composites over the years, despite improvements in mechanical properties, and it can contribute to gap formation at the margins of restorations. The current goal of the manufacturers of resin composite restorative materials remains improvement or elimination of contraction stress—possibly through low or non-shrinking monomers.[17] Research has shown that the amount of polymerization shrinkage may be reduced by an increase in filler particles.[8] Filler loading can be deceiving, however, because it may not be indicative of the presence of pre-polymerized filler particles.[16] With the use of pre-polymerized filler particles, resin composites typically benefit by having lower polymerization shrinkage, as found with Durafill VS and Premise. However, in the current study, it was interesting to note that Gradia Direct Posterior did not benefit from their incorporation.

Although it was not possible to isolate the effect of the additional use of pre-polymerized filler particles on the properties of hybrid resin composites, it may be speculated that a reduction in strength, modulus and microhardness may be the result of their incorporation. Traditionally, mechanical properties, such as strength and stiffness, are generally inferior with resin composite materials containing pre-polymerized particles, such as microfills, because of the lower filler content.[14] The advantages of increased esthetics and polish retention with hybrid resin composites containing pre-polymerized filler particles may be offset by a possible reduction in strength properties, especially in posterior areas.[13] Other factors besides filler content, such as the degree of conversion and type and the ratio of monomers, also affect the mechanical properties of resin composites. However, the filler component is considered the most significant factor in strength properties and, therefore, more often studied.[18] Based on the results of the current study, stronger hybrid resin composite restorative materials are available other than Gradia Direct Posterior and Premise for stress-bearing restorations.

CONCLUSIONS

Within the limitations of the current study, in general, traditional hybrid composites (Z250, Esthet-X) had higher strength and modulus; hybrid composites containing pre-polymerized fillers (Gradia Direct Posterior, Premise) performed more moderately and microfill (Durafill VS) had lower strength. Premise and Durafill VS had the lowest polymerization shrinkage.

Disclosure

The views expressed in this article are those of the authors and do not reflect the official policy of the Department of Defense or other departments of the United States Government. The authors do not have any financial interest in the companies whose materials are discussed in this article.

(Received 9 December 2008)

References

1. Anusavice KJ (2003) *Phillip's Science of Dental Materials* WB Saunders Co, Philadelphia.

2. Ferracane JL (1995) Current trends in dental composites *Critical Reviews in Oral Biology and Medicine* **6(4)** 302-318.

3. Premise Technical Information. Retrieved on June 19, 2008 from: *http://www.kerrdental.com*.

4. Gradia Direct Posterior Technical Information. Retrieved on June 19, 2008 from: *http://www.gcamerica.com*.

5. Mitra SB, Wu D & Holmes BN (2003) An application of nanotechnology in advanced dental materials *Journal of the American Dental Association* **134(10)** 1382-1390.

6. ISO-Standards 4049 (2000) Dentistry—Polymer-based filling, restorative and luting materials *Geneve: International Organization for Standardization*.

7. Rodrigues SA Junior, Ferracane JL & Della Bona A (2008) Flexural strength and Weibull analysis of a microhybrid and nanofill composite evaluated by 3- and 4-point bending tests *Dental Materials* **24(3)** 426-431.

8. Sabbagh J, Ryelandt L, Bacherius L, Biebuyck JJ, Vreven J & Lambrechts P (2004) Characterization of the inorganic fraction of resin composites *Journal of Oral Rehabilitation* **31(11)** 1090-1101.

9. Watanabe H, Khera SC, Vargas MA & Qian F (2008) Fracture toughness comparison of six resin composites *Dental Materials* **24(3)** 418-425.

10. Masouras K, Silikas N & Watts DC (2008) Correlation of filler content and elastic properties of resin-composites *Dental Materials* **24(7)** 932-939.

11. Sabbagh J, Vreven J & Leloup G (2002) Dynamic and static moduli of elasticity of resin-based materials *Dental Materials* **18(1)** 64-71.

12. Ersoy M, Civelek A, L'Hotelier E, Say EC & Soyman M (2004) Physical properties of different composites *Dental Materials Journal* **23(3)** 278-283.

13. Lu H, Lee YK, Oguri M & Powers JM (2006) Properties of a dental resin composite with a spherical inorganic filler *Operative Dentistry* **31(6)** 734-740.

14. Powers JM & Sakaguchi RL (2006) *Craig's Restorative Dental Materials* Mosby, St Louis.

15. Sharp LJ, Choi IB, Lee TE, Sy A & Suh BI (2003) Volumetric shrinkage of composites using video-imaging *Journal of Dentistry* **31(2)** 97-103.

16. Tiba A, Charlton DG, Vandewalle KS & Ragain JC Jr (2005) Comparison of two video-imaging instruments for measuring volumentric shrinkage of dental resin composites *Journal of Dentistry* **33(9**) 757-763.

17. Ferracane JL (2008) Buonocore Memorial Lecture. Placing dental composites—a stressful experience *Operative Dentistry* **33(3)** 247-257.

18. Rodrigues SA Jr, Zanchi CH, Varella de Carvalho R & Demarco FF (2007) Flexural strength and modulus of elasticity of different types of resin-based composites *Brazilian Oral Research* **21(1)** 16-21.