Silorane- and high filled-based "low-shrinkage" resin composites: shrinkage, flexural strength and modulus

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Abstract: This study compared the volumetric shrinkage (VS), flexural strength (FS) and flexural modulus (FM) properties of the low-shrinkage resin composite Aelite LS (Bisco) to those of Filtek LS (3M ESPE) and two regular dimethacrylate-based resin composites, the microfilled Heliomolar (Ivoclar Vivadent) and the microhybrid Aelite Universal (Bisco). The composites (n = 5) were placed on the Teflon pedestal of a video-imaging device, and VS was recorded every minute for 5 min after 40 s of light exposure. For the FS and FM tests, resin discs (0.6 mm in thickness and 6.0 mm in diameter) were obtained (n = 12) and submitted to a piston-ring biaxial test in a universal testing machine. VS, FS, and FM data were submitted to two-way repeated measures and one-way ANOVA, respectively, followed by Tukey's *post-hoc* test (α = 5%). Filtek LS showed lower VS than did Aelite LS, which in turn showed lower shrinkage than did the other composites. Aelite Universal and Filtek LS exhibited higher FS than did Heliomolar and Aelite LS, both of which exhibited the highest FM. No significant difference in FM was noted between Filtek LS and Aelite Universal, while Heliomolar exhibited the lowest values. Aelite LS was not as effective as Filtek LS regarding shrinkage, although both low-shrinkage composites showed lower VS than did the other composites. Only Filtek LS exhibited FS and FM comparable to those of the regular microhybrid dimethacrylate-based resin composite.

Descriptors: Dental Materials; Composite Resins; Polymerization.

Introduction

Despite improvements in the mechanical properties of resin composites (RCs), polymerization shrinkage still remains a challenge and imposes limitations on the clinical use of RCs.[1] Shrinkage is caused by an exchange of van der Waals spaces for shorter covalent bond spaces when monomer molecules are converted into a polymer network.[2] The resulting composite shrinkage of 2%–5%[3,4] generates stress at the dentin/adhesive interface,[5,6] causing cusp deflection[7,8] or de-bonding, marginal staining, enamel cracking, and/or post-operative sensitivity.[9]

Many strategies have been proposed to reduce the shrinkage stress created during the polymerization of RCs into the prepared tooth cavity. Some of these techniques include the use of liners and the incremental placement of composites to allow them to shrink freely toward the adhesive interface.[10] Other strategies focus on slowing down the polymer-

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ization rate to extend the pre-gel phase,[5,11] allowing enough plastic deformation (flow) to compensate for the reduction in RC volume. Such a controlled polymerization can be achieved by initial light exposure with low light intensity followed by a final cure at high intensity (soft-start polymerization)[11] by applying short pulses of light energy (pulse-delay technique)[12] or a combination of both techniques.

Another approach to reduce shrinkage stress is the use of the so-called "low-shrinkage" RCs (LSRCs). To develop LSRCs, manufacturers have replaced monomers such as TEGDMA with monomers with increased molecular weights.[10] As a consequence, RCs have fewer double bonds per unit of weight, leading to less shrinkage during polymerization.[3,13] Another monomer known as silorane has been added to the composition of some RCs.[14] In that system, the ring-opening chemistry of the monomer starts with the cleavage and opening of the ring systems to gain space and to counteract the volume reduction that occurs when the chemical bonds are formed.[14] Most recently, some authors have observed lower shrinkage stress created by the polymerization of a resin composite with nanogel-modified monomer added to its composition.[15]

As another alternative to LSRCs, manufacturers have incorporated high levels of fillers into the resin matrix, resulting in a low resin matrix fraction. Once the resin matrix determines the reduction in volume when the dense cross-linked polymer network is created,[16] these RCs are expected to develop low shrinkage during polymerization. While the silorane-based RC has been exhaustively evaluated regarding its shrinkage values[14,17] and mechanical properties,[18,19] little information is available in the literature concerning the shrinkage and mechanical properties of these highly filled LSRCs.[17]

The analysis of flexural strength (FS) and flexural modulus (FM), along with volumetric shrinkage (VS), is crucial in predicting the clinical success of composites. Review studies have demonstrated that RC fracture is one of the main reasons for restoration failures,[10,20] as LSRCs with low FS are expected to fail prematurely, compromising restoration longevity. Similarly, FM is also closely related to the durability of restorative procedures with RCs because products with low FM show severe elastic deformity under functional stresses, leading to the clinical failure of restorations.[21]

Thus, the aim of the current study was to compare the VS, FS, and FM of one highly filled LSRC with those of a silorane-based LSRC and two regular dimethacrylate-based RCs. The research hypothesis was that the highly filled LSRC presents similar VS as the silorane-based LSRC, with FM and FS comparable to those observed in a regular-dimethacrylate microhybrid RC.

Methodology

Volumetric shrinkage analysis

The VS values of two microhybrid LSRCs and two regular dimethacrylate-based resin composites, one microfilled and another microhybrid RC (Table 1), were measured using a video-imaging device (AccuVol; Bisco Inc., Schaumburg, USA) in the single-view mode. Since the microhybrid dimethacrylate-based RC is recommended for posterior teeth, its mechanical properties were used as a reference for comparison with the mechanical properties of both LSRCs. Conversely, a microfilled RC was selected because manufacturers do not recommend its use on posterior teeth. Therefore, any results from both LSRCs that are close to those from the microfilled RC indicates that their use on posterior teeth should be avoided.

Each specimen (n = 5) was shaped into a semisphere (with volumes averaging approximately 5 μL) and placed on the Teflon pedestal along the light path. The RC was allowed to rest for 1 min and was later exposed to 40 s of light curing (Astrallis 10, power output: 950 mW/cm^2; Ivoclar Vivadent, Schaan, Liechtenstein), with the curing unit tip positioned 8 mm from the specimen. The curing light intensity was constantly measured with a radiometer (Optilux Radiometer model-100 SDS; Demetron Kerr, Danbury, USA). VS (%) was recorded every minute for 5 min after the initiation of light activation, the period during which most shrinkage occurs.[3]

Flexural strength and modulus

The composites were applied into Teflon molds

Table 1 - Materials used in this study.

Product	Composition (batch number)	Composite type / filler content (volume)
Aelite LS (ALS, Bisco Inc., Schaumburg, USA)	Bis-EMA; dental glass; amorphous silica (0600008366)	Microhybrid / 74%
Aelite Universal (AU, Bisco Inc., Schaumburg, USA)	Bis-EMA; TEGDMA; glass filler; amorphous silica (0600009124)	Microhybrid / 53%
Filtek LS (3MLS, 3M ESPE, St. Paul, USA)	Silane treated quartz; yttrium trifluoride; bis-3,4-epoxycyclohexylethyl-phenyl-methylsilane; 3,4-epoxycyclohexylcyclopolymethylsiloxane; mixture of epoxy-mono-silanole by-products; mixture of epoxyfunctional di- and oligo-siloxane byproducts; mixture of alpha-substituted by-products (7AC)	Microhybrid / 55%
Heliomolar (HEL, Ivoclar Vivadent, Schaan, Liechtenstein)	Bis-GMA, UDMA, decamethylendimethacrylate, silicondioxide; ytterbiumtrifluoride, catalysts, stabilizers and pigments (H29947)	Microfilled / 46%

Bis-EMA: ethoxylated bisphenol A dimethacrylate; UDMA: diurethane dimethacrylate; Bis-GMA: bisphenol A diglycidyl ether methacrylate.

to create disc-shaped specimens with dimensions of approximately 0.6 mm thickness and 6.0 mm diameter. Each specimen was covered with a Mylar strip and a microscope glass slide, after which manual pressure was applied to force the material to flow into the mold. The RCs were exposed to light from the same light-curing unit for 20 s on both sides, resulting in a 40-s light exposure. Excess material was removed, and the specimen surfaces were wet ground with 1200- and 2000-grit SiC papers to create flat surfaces and adjust specimen dimensions. As such, the dimensions of all of the specimens were measured with a digital caliper (MDC-Lite, Mitutoyo Corporation; Kanagawa, Japan) after these procedures. The discs (n = 12) were dark-stored in relative humidity for 24 h before the biaxial flexural test was performed.

The discs were individually placed into a custom-made testing jig and tested for biaxial flexure strength using the piston-ring biaxial test on a universal testing machine (Instron 5844, Instron Corp., Canton, USA) at 1.27 mm/min until failure. The maximum load was recorded for each specimen, and the elastic modulus was determined from the linear portion of each stress/strain curve. The following formula for the biaxial flexural strength (σ) was used:

$$\sigma = -0.238 \times 7P(X - Y) / b^2,$$

where
σ is the maximum center tensile stress (megapascals),
P is the total load causing fracture (newtons),
$X = (1 + \upsilon)ln(r_2 / r_3)^2 + [(1 - \upsilon) / 2](r_2 / r_3)^2$,
$Y = (1 + \upsilon)[1 + ln(r_1 / r_3)^2] + [(1 - \upsilon)(r_1 / r_3)^2]$ and
b is the specimen thickness at fracture origin (millimeters),

in which
υ is Poisson's ratio (used $\upsilon = 0.25$),
r_1 is the radius of the support circle (millimeters),
r_2 is the radius of the loaded area (millimeters) and
r_3 is the radius of the specimen (millimeters).

The FS and FM were calculated using SRS Biaxial Testing Software (Instron Corp., Canton, USA) and were expressed in MPa and GPa, respectively. The FS and FM data were normal and homocedastic.

Statistical analysis

The VS values were submitted to two-way repeated measures ANOVA, while the FS and FM data were submitted to one-way ANOVA. Significant differences among the groups of VS, FS, and FM analyses were detected using Tukey's *post-hoc* test (pre-set alpha of 0.05). All of the testing was performed using personal statistical software (SAS 8.0 for Win-

dows; SAS Institute Inc., Cary, USA). *Post-hoc* power analysis was performed to analyze the VS, FS, and FM data using additional software (Statistics 19, SPSS Inc., IBM Company, Armonk, USA).

Results

Post-hoc power analysis demonstrated a statistical power greater than 95% at a pre-set alpha of 0.05 for all tested variables. The LSRCs showed the lowest VS among all of the products (Table 2), while the VS of 3MLS was lower than that of ALS regardless of the time interval ($p < 0.0001$). AU exhibited the highest VS values among all of the products ($p < 0.0001$). For all of the products, most VS occurred within the first minute, followed by a significant increase from 1- to 2-min intervals ($p < 0.001$). No significant difference in VS was observed between 2- and 5-min intervals for any of the products, except for 3MLS, which showed further increase in VS values from 3- to 5-minute intervals ($p = 0.0130$).

Figure 1 exhibits the FS (A) and FM (B) of all of the products. The 3MLS and AU products showed the highest FS values, while the HEL and ALS products showed the lowest values ($p < 0.0001$). ALS showed the highest FM values, while HEL exhibited the lowest FM values. No significant difference in FM was noted between 3MLS and AU, which showed FM values significantly higher than that of HEL and lower than that of ALS ($p < 0.0001$).

Discussion

The current results confirmed that the evaluated LSRCs have lower VS values than do the other RCs, although ALS exhibited higher VS than did 3MLS. Furthermore, only 3MLS exhibited similar mechanical properties to those observed for the regular dimethacrylate-based microhybrid composite, while ALS showed lower FS than did the microhybrid dimethacrylate-based RC. Therefore, the research hypothesis that ALS presents VS similar to that of the silorane-based LSRC, with FM and FS comparable to those of the regular-dimethacrylate microhybrid RC, was rejected.

With regard to the VS of both LSRCs, increased filler content in ALS was not as effective as the inclusion of silorane monomer in 3MLS. Notably, after exposure of 3MLS to light, the polymerization efficiency of the cationic ring-opening monomers

Table 2 - Means and standard deviations of the volumetric shrinkage (%) of RCs based on a 5-min analysis.

	1 min	2 min	3 min	4 min	5 min
Aelite Universal (AU)	2.70 (0.14) Aa	2.97 (0.09) Ab	3.01 (0.10) Ab	3.04 (0.10) Ab	3.04 (0.12) Ab
Heliomolar (HEL)	1.98 (0.09) Ba	2.27 (0.07) Bb	2.32 (0.07) Bb	2.35 (0.07) Bb	2.35 (0.09) Bb
Aelite LS (ALS)	1.45 (0.11) Ca	1.60 (0.08) Cb	1.63 (0.05) Cb	1.65 (0.05) Cb	1.65 (0.10) Cb
Filtek LS (3MLS)	1.02 (0.08) Da	1.27 (0.06) Db	1.33 (0.06) Dbc	1.36 (0.06) Dcd	1.36 (0.07) Dd

Means followed by different letters (capital letters within column; lower case letters within row) are significantly different.

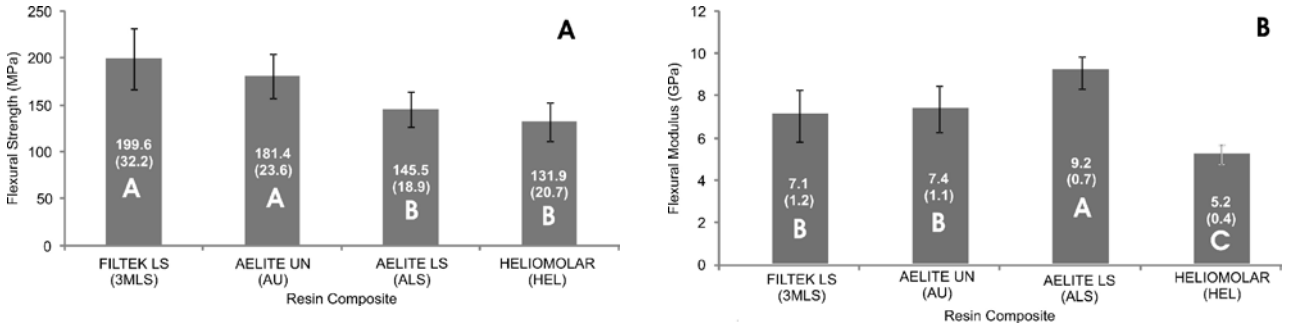

Figure 1 - Bar graphs showing the FS (**A**) and FM (**B**) of the RCs. Different upper case letters within the bars represent significant differences among the means when one-way ANOVA and Tukey's *post-hoc* test were performed at a pre-set alpha of 5%.

increased only after longer periods.[8] As a consequence, silorane-based composites achieved lower monomer conversion than did other RCs within the first minutes after light exposure.[8] These slower polymerization kinetics may have also contributed to the lowest VS observed in 3MLS and also explain why only 3MLS showed continued shrinkage from 3- to 5-minute intervals after light-activation.

ALS exhibited the lowest FS among the values obtained from microfilled HEL, despite its high filler fraction (88% by weight according to the manufacturer). Composites with filler contents greater than 80% by weight have reduced fatigue resistance.[22] Based on Soderholm's theoretical determination of shrinkage stresses in composites,[23] a crack will not form as easily in an RC with low filler content as it will in a highly filled composite because the former displays higher tangential tensile stress. Moreover, increased filler content results in decreased interparticle spacing.[24] Although the stress intensification factor (Kc) increases with a decrease in space until critical spacing is reached, Kc decreases with further filler addition after the critical volume fraction is exceeded,[24] as stress is dissipated on the filler rather than on the resin matrix.[25] As a consequence, some mechanical properties, such as tensile strength, may be compromised, as was observed in the current study.

On the other hand, 3MLS, along with AU, also exhibited the highest FS values among all composites. FS is related to the polymer type and filler content regarding filler distribution and orientation.[26] 3MLS has quartz particles, whose spatial orientation can be described as a crystalline solid network of interconnected SiO_4 tetrahedra and classified as tectosilicate. Conversely, the other tested materials consisted predominantly of glass, whose silica (SiO_2) structures have an amorphous (non-crystalline) orientation.[27] Such differences in filler composition do not allow a reliable analysis of the effects of filler size and shape on mechanical properties of commercial RCs. Moreover, because of the differences in filler composition among the products, it was not possible to distinguish the influence of silorane monomer from the influence of the filler particle features on the FS observed in 3MLS.

The current study evaluated the VS of LSRCs and RCs over a 5-min period. Although most of the composite shrinkage was observed within this interval,[3] further shrinkage is expected as monomer conversion continues over a period of 24 h.[28] For this reason, the VS values reported in this study may not represent the total shrinkage of each RC. Moreover, all of the mechanical properties observed in the evaluated LSRCs cannot be extrapolated to other commercially available LSRCs, as differences in monomer and filler composition among products may result in better or worse mechanical properties than those observed in the evaluated LSRCs. Therefore, further studies evaluating LSRCs with other compositions, as well as with different total shrinkage, are required.

Conclusion

Within the limitation of this study, and despite its lower VS than those of regular dimethacrylate based-resin composites, ALS exhibited higher volumetric shrinkage than did 3MLS. Only 3MLS showed mechanical properties comparable to those of regular dimethacrylate-based microhybrid composite.

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References

1. Ferracane JL. Resin-based composite performance: are there some things we can't predict?. Dent Mater. 2013 Jan;29(1):51-8.
2. Peutzfeldt A. Resin composites in dentistry: the monomer systems. Eur J Oral Sci. 1997 Apr;105(2):97-116.
3. Naoum SJ, Ellakwa A, Morgan L, White K, Martin FE, Lee IB. Polymerization profile analysis of resin composite dental restorative materials in real time. J Dent. 2012 Jan;40(1):64-70.

4. Lee IB, Cho BH, Son HH, Um CM. A new method to measure the polymerization shrinkage kinetics of light cured composites. J Oral Rehabil. 2005 Apr;32(4):304-14.
5. Feilzer AJ, De Gee AJ, Davidson CL. Quantitative determination of stress reduction by flow in composite restorations. Dent Mater. 1990 Jul;6(3):167-71.
6. Boaro LC, Goncalves F, Braga RR. Influence of the bonding substrate in dental composite polymerization stress testing. Acta Biomater. 2010 Feb;6(2):547-51.
7. Lee MR, Cho BH, Son HH, Um CM, Lee IB. Influence of cavity dimension and restoration methods on the cusp deflection of premolars in composite restoration. Dent Mater. 2007 Mar;23(3):288-95.
8. Palin WM, Fleming GJ, Nathwani H, Burke FJ, Randall RC. In vitro cuspal deflection and microleakage of maxillary premolars restored with novel low-shrink dental composites. Dent Mater. 2005 Apr;21(4):324-35.
9. Gaengler P, Hoyer I, Montag R, Gaebler P. Micromorphological evaluation of posterior composite restorations - a 10-year report. J Oral Rehabil. 2004 Oct;31(10):991-1000.
10. Ferracane JL. Resin composite--state of the art. Dent Mater. 2011 Jan;27(1):29-38.
11. Silikas N, Eliades G, Watts DC. Light intensity effects on resin-composite degree of conversion and shrinkage strain. Dent Mater. 2000 Jul;16(4):292-6.
12. Kanca J, 3rd, Suh BI. Pulse activation: reducing resin-based composite contraction stresses at the enamel cavosurface margins. Am J Dent. 1999 Jun;12(3):107-12.
13. Yap AU, Soh MS. Post-gel polymerization contraction of "low shrinkage" composite restoratives. Oper Dent. 2004 Mar-Apr;29(2):182-7.
14. Weinmann W, Thalacker C, Guggenberger R. Siloranes in dental composites. Dent Mater. 2005 Jan;21(1):68-74.
15. Moraes RR, Garcia JW, Barros MD, Lewis SH, Pfeifer CS, Liu J, et al. Control of polymerization shrinkage and stress in nanogel-modified monomer and composite materials. Dent Mater. 2011 Jun;27(6):509-19.
16. Kleverlaan CJ, Feilzer AJ. Polymerization shrinkage and contraction stress of dental resin composites. Dent Mater. 2005 Dec;21(12):1150-7.
17. Boaro LC, Goncalves F, Guimaraes TC, Ferracane JL, Versluis A, Braga RR. Polymerization stress, shrinkage and elastic modulus of current low-shrinkage restorative composites. Dent Mater. 2010 Dec;26(12):1144-50.
18. Papadogiannis D, Tolidis K, Lakes R, Papadogiannis Y. Viscoelastic properties of low-shrinking composite resins compared to packable composite resins. Dent Mater J. 2011 30(3):350-7.
19. Leprince J, Palin WM, Mullier T, Devaux J, Vreven J, Leloup G. Investigating filler morphology and mechanical properties of new low-shrinkage resin composite types. J Oral Rehabil. 2010 May 1;37(5):364-76.
20. Sarrett DC. Clinical challenges and the relevance of materials testing for posterior composite restorations. Dent Mater. 2005 Jan;21(1):9-20.
21. Ilie N, Hickel R. Investigations on mechanical behaviour of dental composites. Clin Oral Investig. 2009 Dec;13(4):427-38.
22. Htang A, Ohsawa M, Matsumoto H. Fatigue resistance of composite restorations: effect of filler content. Dent Mater. 1995 Jan;11(1):7-13.
23. Soderholm KJ. Influence of silane treatment and filler fraction on thermal expansion of composite resins. J Dent Res. 1984 Nov;63(11):1321-6.
24. Lloyd CH, Iannetta RV. The fracture toughness of dental composites. I. The development of strength and fracture toughness. J Oral Rehabil. 1982 Jan;9(1):55-66.
25. Cross M, Douglas WH, Fields RP. The relationship between filler loading and particle size distribution in composite resin technology. J Dent Res. 1983 Jul;62(7):850-2.
26. Masouras K, Silikas N, Watts DC. Correlation of filler content and elastic properties of resin-composites. Dent Mater. 2008 Jul;24(7):932-9.
27. Lien W, Vandewalle KS. Physical properties of a new silorane-based restorative system. Dent Mater. 2010 Apr;26(4):337-44.
28. Rueggeberg FA, Caughman WF. The influence of light exposure on polymerization of dual-cure resin cements. Oper Dent. 1993 Mar-Apr;18(2):48-55.