Long-term Volumetric Analysis of Composites Resins Stored in PBS

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Volumetric Evaluation of recently Introduced Bulk-Fill Restorative Material

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Abstract

Objective: To evaluate the volumetric change (polymerization shrinkage/hygroscopic expansion) of 4 bulk-fill restorative composites and 2 incremental/conventional fill composites over 60 days of storage in 0.2M physiologic phosphate buffered saline (PBS).

Methods: Composite specimens (n=15) were created on keyed pedestals following manufacturer recommendations, and stored in PBS at 37 °C until the appointed testing time. Volumetric measurements were recorded using a video imaging analysis instrument (AcuVol, BISCO Inc.). Volumetric measurements were captured at following intervals: pre-polymerized, immediate post-polymerized, 1, 3, 7, 14, 21 and 30 days. Statistical analysis: Friedman with post stepwise step-down test ($p = 0.05$) and Kruskal-Wallis with post stepwise step-down test ($p = 0.05$).

Results: All materials underwent significant polymerization shrinkage from their pre-polymerized volumes; conventional composites peaked at day 1, bulk-fill on day 3. ACTIVA BioACTIVE RESTORATIVE, Filtek One Bulk Fill Restorative, Filtek Supreme Ultra (FSU), and Revolution 2 returned to a volume statistically similar to their pre-polymerized volume. SonicFill 2 demonstrated expansion, but not statistically similar to its pre-polymerized volume. Fill-Up! did not show significant expansion from peak shrinkage. FSU showed earliest hygroscopic expansion, statistically similar to pre-polymerized volume by day 3.

Conclusion: No composite resin was dimensionally stable and all absorbed water. Fill-Up! underwent high and unreversed shrinkage. More research is needed to establish the clinical impact of these findings.

Manuscript

Introduction

There has been a paradigm shift towards digital dentistry and milled ceramics. These workflows benefit from fast setting, tooth colored, and bondable core build up material like composite resin. Traditional materials such as Amalgam or cast metal are unaesthetic and do not facilitate same day crown production. Furthermore, Amalgam use is being phased down. The Minamata convention goal of amalgam phase out looms in 2020. With the added benefit posterior and even anterior restorative indications, composite is becoming the go dental material.

Composite is not a perfect dental material. Polymerization shrinkage is found in virtually all composite formulations to some degree. Breakage of carbon double bonds produces carbon polymer chains with close molecular proximity (1). This causes molecular contraction. The prolonged action of the shrinkage has not been established, though composite materials do shrink 1.35% to 7.1% (1). A number of factors influence polymerization shrinkage. Some are degree of conversion, resin matrix constituents, and number of bonded walls (2).

Dimensional stability is described as a desirable property for core materials (3). Dimensional changes of a variety of light cured bulk fill, traditional, and dual cure composite resins indicated for build ups and restorative were investigated. A video imaging device (AcuVol, BISCO Inc.) was used to record volumetric changes. This study investigated: 1.) Is there a difference in polymerization shrinkage between these composite materials? 2.) Is there a difference in hygroscopic expansion between these composite materials? The hypothesis is that there is no difference.

Methods and Materials

Samples:

Ninety keyed pedestals produced for AcuVol by BISCO were prepared. The pedestal's bonding surface was roughened to allow mechanical retention. A single layer of bond (Optibond All-in-One, Kerr) was placed on bonding surface and light cured for 15 seconds. The pedestal was blacked out with black nail polish to allow for easier differentiation of samples and consistent, repeated measures.

Hemispherical samples were formed on pedestals. 6 groups (n=15) prepared. Polymerization was accomplished utilizing a PolyWave® visible curing light (BluePhase G2, Ivoclar Vivadent, Amherst, NY, USA). Irradiance periodically verified to be >1100 mW/cm2 using radiometer (Bluephase Meter II, IvoclarVivadent, Amherst, NY, USA).

Groups:

Filtek One Bulk Fill Restorative	3M ESPE, St. Paul, MN, USA	Light cured bulk-fill composite	Bis-GMA, bis-EMA, UDMA, zirconia, Filler load:76.5 wt%, 58.4 vol%.
Fill-up!	Coltène/Whaledent AG	Dual curing bulk composite	TMPTMA, UDMA, bis-GMA, TEGDMA, dibenzoyl peroxide; benzoyl peroxide, Zinc oxide coated. Filler load: 65 wt%; 49 vol%
Filtek Supreme Ultra	3M ESPE, St. Paul, MN, USA	Light cured composite	bis-GMA, UDMA, TEGDMA, and bis-EMA(6) resins 78.5% by weight(63.3% by volume)
SonicFill 2	Kerr Corporation, CA, USA	Light cured bulk-fill composite	Bis-GMA, TEGDMA, Bis-EMA. Filler load: 81.3% wt % unreported.
Revolution 2	Kerr Corporation, CA, USA	Light cured flowable composite	Bis-GMA, TEGDMA RF material contained barium glass and synthetic silica fillers (55% by weight) with particle size of 1μm
ACTIVA BioACTIVE-RESTORATIVE	Pulpdent Corp., Watertown, MA, USA	Bioactive Restorative Glass	Blend of diurethane and other methacrylates with modified polyacrylic acid (44.6%), amorphous silica (6.7%), and sodium fluoride (0.75%)

Volumetric Change Determination:

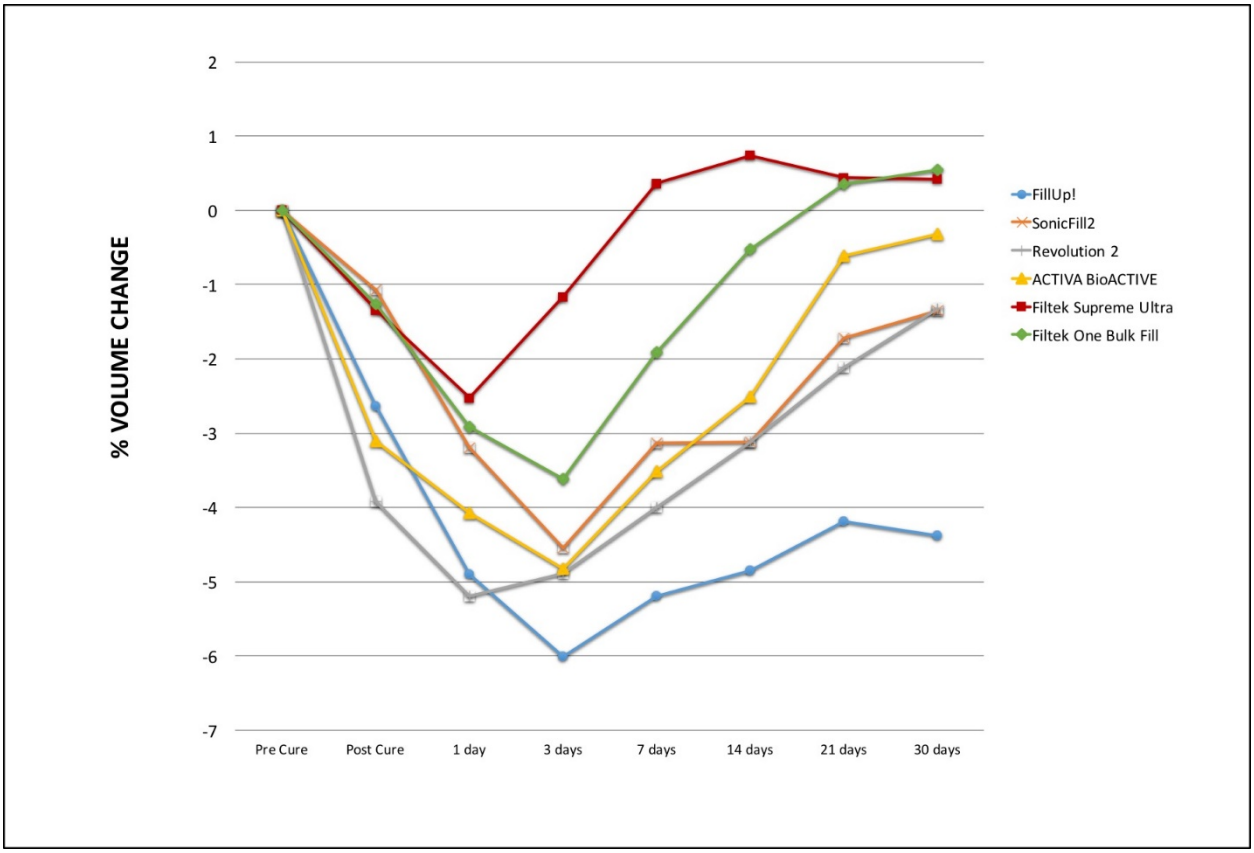
AcuVol video imaging was used to compare sample volume using single view volumetric reconstruction mode. Single and multi-view reconstruction modes have been shown to give reproducible results for volume reconstruction values (4). Composite specimens were stored in (0.2 M) phosphate buffered solution (PBS) at 37 °C until the appointed testing time. PBS selected as it is isotonic and mimics salivary pH. A blacked is pedestal shown below. Volumetric measurements (in ml) were captured at following intervals: pre & post-polymerization, 1, 3, 7, 14, 21, and 30 days.

Statistical Analysis:

Data determined to be non-parametric by Levene and Shapiro Wilk's tests, and non-parametric analysis was conducted. A repeated measures test was used to compare each composite group over time; Friedman's test with Dunnett's post hoc test. Comparisons between composite groups were accomplished using Kruskal-Wallis test with Dunn's pairwise post hoc.

All of tests were at a 95 percent level of confidence ($\alpha = 0.05$). Statistical analysis was performed using SPSS 21 (IBM/SPSS, Chicago, IL, USA).

Results



The percent pre-cure volume for each group is shown above. All groups underwent significant polymerization shrinkage. An initial period of shrinkage (approx. 2-6 percent) is apparent within the first 3 days. Hygroscopic expansion is observed for 30 days. No sample underwent significant changes in volume between 21 days and 30 days. In most groups, polymerization shrinkage was reversed by day 30.

% Volume Change								
Groups	Pre Cure	Post Cure	24 Hours	3 Days	7 Days	14 Days	21 Days	30 Days
Filtek Supreme	0% AB	-1.34% (0.3) C a	-2.53% (1.8) C a	-1.16% (2.2) BC a	0.37% (2) AB a	0.74% (1.5) A a	0.44% (1.1) AB a	0.42% (1.3) AB a
Revolution2	0% A	-3.92% (0.7) BCD d	-5.19% (1.3) D b	-4.88% (3.1) CD bc	-4.00% (3.2) BCD bc	-3.12% (2.9) BC cd	-2.11% (3.2) AB b	-1.32% (1.9) A b
Filtek One	0% AB	-1.27% (0.4) CD a	-2.91% (1.0) E a	-3.61% (1.4) E b	-1.91% (1.4) D b	-0.53% (0.8) BC b	0.35% (1) AB a	0.55% (1) A a
SonicFill2	0% A	-1.08% (0.2) B a	-3.20% (1.1) C a	-4.54% (1.1) D bc	-3.13% (1.2) C bc	-3.11% (1.6) C cd	-1.72% (1.4) BC b	-1.35% (1.9) B b
ACTIVA	0% A	-3.10% (0.5) BC c	-4.08% (0.9) CD b	-4.82% (1.5) D bc	-3.51% (1.2) C c	-2.51% (1.4) B c	-0.61% (0.7) A b	-0.32% (1) A ab
Fill-Up!	0% A	-2.63% (0.6) B B	-4.90% (1.7) C b	-6.01% (1.7) D c	-5.19% (1.5) CD d	-4.85% (1.9) C d	-4.19% (1.9) BC c	-4.38% (1.7) C c

n=15; Capital letters annotate statistically similar groups per row (Friedman Test with post stepwise step-down test)
Lowercase letters annotate statistically similar groups per column (Kruskal-Wallis Test with post stepwise step-down test)
Pre Cure is baseline (0% vol change); Negative values indicate shrinkage/reduced volume from baseline; positive values indicate volume larger than baseline

Conventional composites (Filtek Supreme Ultra and Revolution 2) underwent peak polymerization shrinkage at 24 hours. Bulk full composites (ACTIVA BioACTIVE-RESTORATIVE, Filtek One Bulk Fill Restorative, SonicFill 2, and Fill-Up!) underwent peak polymerization at day 3. With the exception of Fill-Up! And SonicFill 2 all groups returned to their pre-polymerized volume.

Fill-Up! underwent the highest peak expansion recorded. Fill-Up! showed only slight expansion following peak shrinkage and was the only group to not return to its post polymerization volume. Only Filtek Supreme Ultra and Filtek One Bulk Fill underwent significantly less peak polymerization shrinkage compared to Fill-Up!

Discussion

Filtek Supreme Ultra and Filtek One Bulk Fill Restorative had minimal polymerization shrinkage which fully reversed. Both are nano composites. Their high filler loading minimizes the influence of resin component on polymerization shrinkage (2). However the actual filler type seems to have little impact on hygroscopic behavior (5). Hydrophobicity of the resin matrix is indicative of the amount of water absorbed by dental composites (6). BisGMA is the typical

backbone for composite formulations. It is hydrophilic, high molecular weight, and highly viscus (6). TEGMA is a diluent molecule able to lower viscosity of BisGMA improve degree of conversion. It is found in most of composite formulations, including Revolution 2, which also contains BisGMA. Revolution 2 had the highest immediate post polymerization shrinkage but also underwent a high degree of hygroscopic expansion which fully return it to pre polymerized volume. Both Filtek formulations and contained relatively hydrophobic BisEMA, which may have resulted in a less hydrophilic resin matrix. Filtek Supreme Ultra and Filtek One Bulk Fill Restorative were able to fully reverse their minimal shrinkage without uncontrolled expansion.

Fill-Up! shrunk significantly more than either Filtek formulation, and had limited hygroscopic expansion. This behavior may be due to Fill-Up!'s relatively novel cross linking agent TMPTMA. It is widely used outside of the dental industry to increase polymer mechanical strength and resistance, although it appears to only be recently introduced to dentistry. TMPTMA is unique to Fill-Up! among composites tested. Its unique properties as a cross linking agent lowers the solubility of dental composites while improving mechanical properties (7). It may explain the lack of hygroscopic expansion and high shrinkage seen in Fill-Up!.

Shrinkage appeared to continue into the post gel phase. Light curing is followed by a dark reaction process. The photopolymerization rapidly initiates the setting of a polymer matrix. However, not all monomers react and may leach into the aqueous environment (8). Following polymerization, free radicals continue to diffuse throughout the vitrified resin matrix. Changes in degree of cure and mechanical properties are seen up to a week. Interestingly, at high curing doses, approximately 2 days is required for peak mechanical properties (9). This time period is greatly increased (7 days) in samples receiving lower curing doses (9). Continued shrinkage during the first 24 hour post curing has been demonstrated (10). The effect of PBS on early

volumetric shrinkage, specifically leaching of monomers and dissolution of free radicals could have an effect on physical volume.

Operator technique plays a role in degree of sorption; it is theorized that the presence of air voids in restorations may create internal zones of oxygen inhibition layers, thus increasing water retention (11). Difficulty shaping SonicFill 2 in open space when forming hemispherical samples may have led to access air voids in the material and a loss of homogeneity of samples, thus effecting the results. More research is needed to ascertain clinical applications and behavior.

The complex interaction between volumetric shrinkage, micro leakage, and polymerization stress has been heavily studied in the literature. Mass gain due to hygroscopic expansion is correlated with relaxation of shrinkage stress (12). However, higher degrees of sorption are correlated with poor color stability (2). A reduction in both hardness and strength is seen when comparing dry verses wet composite (13). Uncontrolled expansion and a reduction in strength have been demonstrated. It has been suggested that excessive sorption and lack of dimensional stability of composite core material could prevent crown seating (14).

On the other hand, residual stresses are generated when polymerization shrinkage is constrained (1). Composite modulus of elasticity and C factor modify these residual stresses. Shrinkage stress can lead to marginal gaps, micro-cracking, marginal staining, cuspal flexion, and sensitivity (1). Composites that can relieve shrinkage stress via hygroscopic expansion may prove to possess a greater potential for clinical success in certain cases. Many of the composites tested, with the exception of Fill-Up!, demonstrated expansion. With microhybrid composites showing up to 97% success rate for class II restorations over 10 years, it appears that hygroscopic expansion and polymerization shrinkage are very well tolerated according to Lempel et al., 2015.

The merits of hygroscopic expansion are unclear. Resin materials undergo both contraction and expansion due to polymerization and water sorption as suggested by this study. According to ISO 4049, the standard for water sorption of polymer based materials is 40 ug/mm3. Even within this standard, the response to the dynamic mouth environment various widely between dental polymers (15).

For same day workflows, this is unlikely to compromise crown seating. However, it has been suggested that hygroscopic expansion of core material can result in inaccurate dies (16). Considering that currently available cements are soluble and do not provide continual impermeability at tooth-cement interface, interim crowns likely will not fully seal a composite core (17). More clinical studies are required to determine if hygroscopic expansion is a source of inaccuracy in fixed prosthodontics. Considering the high clinical success rate of both composite cores and restorations it appears that lack of dimensional stability is generally not problematic. The clinical impact of high shrinkage and low hygroscopic expansion composites resins such as Fill-Up! remains to be determined.

Conclusion

No composite resin was dimensionally stable and all absorbed water. Fill-Up! underwent high and unreversed shrinkage. More research is needed to establish the clinical impact of these findings.

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