ORIGINAL PAPER

DO HIGH-VISCOSITY BULK-FILL COMPOSITES POLYMERIZE SUFFICIENTLY AT DEEP LAYERS?

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ABSTRACT

Introduction: High-viscosity bulk-fill composites may polymerize insufficiently at deep layers after a short curing time.

Objectives: This *in-vitro* study aimed to estimate and compare the degree of conversion (DC) and micro-hardness (MH), following 20 and 40 seconds photopolymerization of three high-viscosity bulk-fill composites at 2-4 mm depth.

Material and methods: Three new brands of bulk-fill dental composites were selected for this study, including Beautifil-Bulk (Shofu Inc., Kyoto, Japan), Tetric N-Ceram Bulk Fill (Ivoclar Vivadent AG, Schaan, Liechtenstein), and Opus Bulk Fill (FGM, Joinville, SC, Brazil). For each composite, six disc samples located at a 2-4 mm depth were prepared via photopolymerization for 20 (n = 3) or 40 (n = 3) seconds. Each sample was 2 mm thick and 8 mm wide. Following one-hour post-cure, DC and MH assessments were applied using Fourier transform infrared spectroscopy (FTIR) and Vickers MH devices. Data were analyzed using parametric ANOVA and LSD tests.

Results: Values of DC and MH were higher following 40 seconds compared with 20 seconds photopolymerization. However, none of the composites reached the minimum accepted DC (≥ 55%). For both curing times, Tetric N-Ceram showed the highest DC% (42%, 51%), while Beautifil-Bulk had the highest MH values for top (58.2 ± 0.5, 60.0 ± 1.5) and bottom (47.5 ± 3.1, 53.5 ± 1.0) surfaces.

Conclusions: At a 2-4 mm depth and one-hour post-cure, bulk-fill composites showed insufficient DC and low MH values when photopolymerized for 20 or 40 seconds. The extension of curing time to 40 seconds improved the DC and MH values.

Key words: bulk-fill composite, composite micro-hardness, composite degree of conversion.

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INTRODUCTION

Even though composites present higher failure rates and increased risks of secondary caries development than amalgam [1], the trend for composite restoration placement in posterior teeth has increased dramatically in the last 10 years [2]. The photopolymerization process of light-cured composites forms a rigid cross-linked polymer matrix via reactivity of the monomer's carbon double bonds [3]. However, some of these carbon bonds remain unreacted (unpolymerized) in the composite restoration [4]. Inadequate polymerization, also referred to as a low degree of conversion (DC) of monomer to polymer, substantially influences mechanical and physical

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properties of the composite, and decreasing the restoration's surface micro-hardness [5]. In addition, low DC increases water sorption, solubility [6, 7], and composite discoloration [8].

Curing light intensity and DC decrease, as composite thickness increases [9, 10]. To overcome the consequences of light attenuation and inadequate polymerization in deep posterior cavities, an incremental technique has been indicated. The method involves a maximum of a 2 mm thick increment and 40 seconds light curing time to achieve sufficient polymerization of composite restoration [11]. However, this technique can lengthen placement time and may increase treatment cost. Recently, manufacturers have developed high-viscosity bulk-fill composites, which could be placed in a single increment up to 4-5 mm thick. Manufacturers have claimed that these bulk-fill composites have the advantage of little attenuation effect on the transmitted curing light. Such an advantage may enable a sufficient DC at deep layers even with a shorter curing time (20 seconds). Therefore, the assessment of polymerization behaviors of new bulk-fill composites appears to be essential to guarantee their potential success in clinical applications.

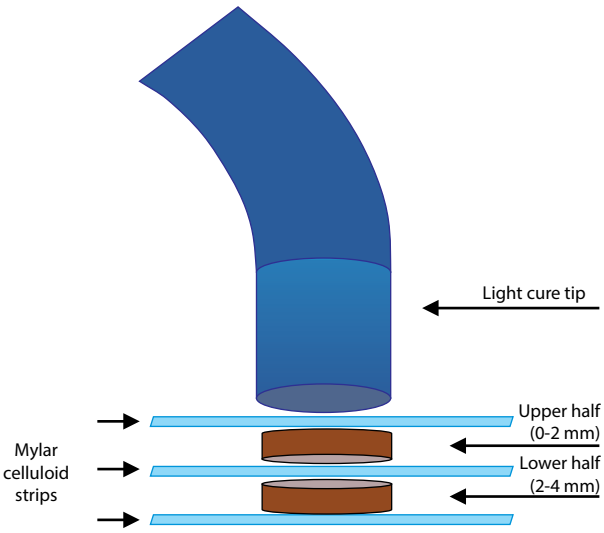

FIGURE 1. Composite sample preparation. The sample consisted of two halves 2 mm thick and 8 mm wide. The upper half was discarded, and the lower half was kept for degree of conversion and micro-hardness assessments

OBJECTIVES

The aim of this *in-vitro* study was to estimate and compare the DC and micro-hardness (MH) of three commercially available bulk-fill composites at a 2-4 mm increment depth, following 20 and 40 seconds of light curing. The null hypothesis was that the two curing times produce no differences between DC or MH values of the tested composites.

MATERIAL AND METHODS

Three new brands of bulk-fill dental composite were selected for this study, including Beautifil-Bulk (Shofu Inc., Kyoto, Japan), Tetric N-Ceram Bulk Fill (Ivoclar Vivadent AG, Schaan, Liechtenstein), and Opus Bulk Fill (FGM, Joinville, SC, Brazil) (Table 1). For each composite, uncured materials were inserted to slightly fill a cylindrical hole mold made of a silicon impression material. The mold comprised two halves of 2 mm height and 8 mm internal diameter assembled on top of each other. At first, the lower half was filled with a composite, followed by the upper half. Care was taken to avoid air entrapment during the composite placement. Three transparent polyester Mylar strips (0.05 mm thick) were placed on the top, bottom, and between mold halves to block the oxygen layer and create a smooth composite surface. The access composite was extruded using a glass slide with firm finger pressure on the top surface. The mold was then irradiated from the top surface (Figure 1) with a new light-emitting diode curing unit (Woodpecker, Guangxi, China) using standard curing mode (1,200 Mw/cm^2). The tip of the light cure device was stabilized in direct contact on the Mylar strip. Six samples were prepared for each composite. Three samples were cured for 20 or 40 seconds according to sample grouping. After polymerization, the upper halves of the composite samples were discarded, whereas the lower halves were kept for study assessments. The lower halves were stored dry in a lightproof container at room temperature for one hour before assessments. For each composite, an uncured sample was used as a control for DC estimation.

A Fourier transform infrared spectrometer (FTIR, Bruker, Ettlingen, Germany) was applied to estimate DC. Each sample was mounted on FTIR stage, and an assessment was undertaken using the following setting: a wavelength of 500-4,000 cm^{-1}, 32 scans, and a resolution of

TABLE 1. Criteria of dental composites used in this study

Composites	Code	Shade	Filler content	Resin matrix
Beautifil-Bulk	PN2034	Universal	(87% weight) fluoro-boro-aluminosilicate	Bis-GMA, Bis-MPEEP, TEGDMA
Tetric N-Ceram Bulk Fill	Z01JJT	Universal	(75-77% weight) barium glass, prepolymer, ytterbium trifluoride, and mixed oxide additives	Dimethacrylates
Opus Bulk Fill	19368	A1	(79% weight) silanized silicon dioxide (silica)	Urethane-dimethacrylates

TABLE 2. Descriptive statistics of degree of conversion percentages (DC%) for tested composites

Groups	Curing time	Minimum	Maximum	Mean	SE
Beautifil-Bulk	20 s	11%	29%	18%	5.50
	40 s	30%	34%	31%	1.37
Tetric N-Ceram Bulk Fill	20 s	18%	50%	42%	9.26
	40 s	37%	61%	51%	7.10
Opus Bulk Fill	20 s	13%	15%	13%	0.61
	40 s	18%	21%	19%	1.03

SE – standard error

6 cm^{-1}. For cured and uncured samples, a ratio of absorbance spectrum peaks for the remaining aliphatic (C = C) and aromatic (C – C) carbon double bonds was estimated at 1,638 and 1,608 cm^{-1}, respectively. DC percentage was then calculated using the following equation:

$$DC\% = 1 - \frac{(1{,}638\ cm^{-1}/1{,}608\ cm^{-1})\ \text{cured sample}}{(1{,}638\ cm^{-1}/1{,}608\ cm^{-1})\ \text{uncured sample}} \times 100$$

Following the DC test, the same samples were subjected immediately to surface MH test using Vickers hardness indenter (Laryee, Beijing, China). A diagonal diamond indenter was subjected to a fixed force of 300 g for a 15 s dual time. Three indentations at a 0.5 mm distance were created in the middle of the top and bottom surfaces. The mean Vickers hardness number of the three indentations was recorded as MH values for each surface.

SPSS statistics version 21 software (IBM, Armonk, USA) was applied to analyze the data. Shapiro-Wilk test was used to investigate the normality of data distribution, and LSD test was used to compare the significance between means ($p < 0.05$). Spearman correlation test was applied to reveal the influence of DC% on MH of the top and bottom surfaces.

RESULTS

Data analyses with Shapiro-Wilk test showed that both DC% and MH were normally distributed around their statistical means. The mean DC% for the tested composites are listed in Table 2. Generally, the 40 seconds curing time produced higher DC% values compared with the 20 seconds curing time, but these differences were not significant ($p \geq 0.05$). For both the curing times, Tetric N-Ceram composite showed the highest DC%, followed by Beautifil-Bulk and Opus Bulk Fill composites. The differences were statistically significant (Table 3), except for Beautifil-Bulk and Opus Bulk Fill, in which their results revealed non-significant differences ($p \geq 0.05$).

The MH means of the top and bottom surfaces are presented in Table 4. Following 40 seconds light curing time, all the composites produced significantly higher MH values compared with 20 seconds light curing time. Among the composites, Beautifil-Bulk had the highest MH values of both surfaces, followed by Tetric N-Ceram and Opus Bulk Fill. The differences were significant, except for a non-significant difference found between the Beautifil-Bulk and Tetric N-Ceram top surfaces. In addition, the composites showed significantly higher MH values of the top surfaces than the bottom surfaces. However, for Beautifil-Bulk, the differences between the top and bottom surfaces were non-significant (Table 3).

TABLE 3. Statistical alpha (α) values for group comparisons in degree of conversion percentages (DC%) and micro-hardness (MH) analysis

Groups/Curing time	DC%	MH (top surface)	MH (bottom surface)
Beautifil-Bulk vs. Tetric N-Ceram Bulk			
20 s	0.045	0.019	0.000
40 s	0.022	0.296	0.000
Beautifil-Bulk vs. Opus Bulk Fill			
20 s	0.539	0.000	0.000
40 s	0.134	0.000	0.000
Tetric N-Ceram Bulk vs. Opus Bulk Fill			
20 s	0.014	0.000	0.000
40 s	0.001	0.000	0.000

Pearson r test revealed a significant ($\alpha = 0.042$) positive correlation ($r = 0.484$) between DC and MH of the top surfaces, and a non-significant ($\alpha = 0.127$) positive correlation ($r = 0.374$) between DC and MH of the bottom surfaces (Figure 2).

DISCUSSION

To simplify the placement procedure, dental companies produce different brands of high-viscosity bulk-fill composites. Companies have assumed that these composites can be placed in a single increment up to 5 mm thick using 20 seconds of curing light. Due to the limit-

ed translucency of composites, previous studies found that an increment of a maximum of 2 mm thick and a minimum of 40 seconds of irradiation time, are clinical requirements for conventional composite restorations [12, 13]. Therefore, the main challenge for the high-viscosity bulk-fill composites is achieving an effective polymerization at a depth greater than 2 mm for a shorter curing time.

This *in-vitro* study assessed DC% at deep layers of three current high-viscosity bulk-fill posterior composite materials irradiated for 20 and 40 seconds curing times. FTIR analytical tool was used to measure DC%, because this tool provides a direct measurement of the amount of unreacted carbon double bonds in polymerized composite [14]. Furthermore, the study also evaluated the potential change in surface MH of composites relative to changes in DC.

Papadogiannis *et al.* suggested that a minimum conversion of 55% is required for the successful application of bulk-fill composites [15]. However, none of the tested composites achieved clinically accepted DC% for both curing times. This result agrees with findings of Jain *et al.* [16]. At deep layers, previous evidence was found of light attenuation that significantly affected the degree of conversion of particular bulk-fill composites [17, 18]. However, the present study results showed that a 40 seconds curing time produced a significant improvement in DC% compared with 20 seconds curing time. This observation agrees with findings of Ajaj *et al.*, who noted that extending the curing time may permit for more free carbon bonds to be involved in polymerization process [19].

Following 1-hour post-cure, the results of this study revealed a much lower DC% than other studies, which used a 24-hour post-cure time interval [17, 20]. Alshali *et al.* found that at 24 hours' post-curing time, DC increased from 15-25% compared with immediately post-cure DC [21]. In the current study, Opus Bulk Fill had the lowest DC%, followed by Beautifil-Bulk, whereas Tetric-N Ceram had the highest DC%. The variation in monomer viscosities of the tested composites can be attributed to the differences in DC%. Monomers with higher viscosities, such as Bis-GMA, have a lower DC% than monomers with lower viscosities, including UDMA and TEGDMA [22, 23]. The low viscosity monomers have more flexible carbon bonds, which increase the monomer reactivity during photopolymerization [23]. Furthermore, the tested composites have different filler types and filler loads. Such differences in filler composition produce a variable influence on the amount of curing light passing through composite samples [24].

We found that extending the curing time from 20 to 40 seconds significantly improved the MH values on both surfaces. In addition, all tested composites showed low mean MH values on the bottom compared with top surfaces, particularly following 20 seconds photopolymerization (MH range, 11.4-47.5). This observation is consistent with the results of de Mendonça *et al.*

TABLE 4. Means ± SE of micro-hardness values for top and bottom surfaces of composite samples

Groups/Curing time	Top	Bottom
Beautifil-Bulk		
20 s	58.2 (± 0.5)	47.5 (± 3.1)
40 s	60.0 (± 1.5)	53.5 (± 1.0)
Tetric N-Ceram		
20 s	52.3 (± 0.4)	34.2 (± 3.0)
40 s	57.4 (± 1.4)	42.0 (± 1.3)
Opus Bulk Fill		
20 s	34.8 (± 1.8)	11.4 (± 1.4)
40 s	42.1 (± 0.6)	21.4 (± 1.1)

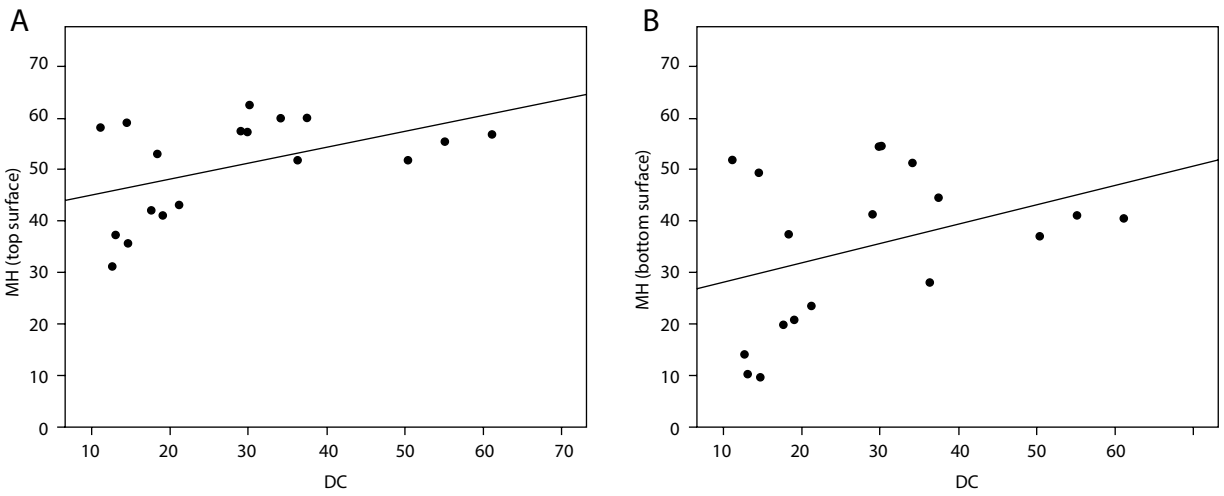

FIGURE 2. Scatter plots with linear fitting lines demonstrate positive correlations between degree of conversion (DC) and micro-hardness (MH) of top and bottom surfaces

study [25]. The low MH values noted in the present work can be attributed to the low DC values. Previous findings showed that MH is a direct representation of DC [26, 27], because unreacted monomers can negatively affect the strength of composites, as confirmed by the positive correlation between DC and MH found in the current study. The Beautifil Bulk composite showed the highest mean MH values on both surfaces compared with the other two composites. Properties, such as translucency of composite material and matching of refractive indices between monomer and filler contents, positively affect DC and therefore, MH of composite samples [24]. Composites with high translucency and matched refractive indices produce little attenuation and scattering of the transmitted light through polymerizing composite [24, 28]. For clinical relevance, manufacturer claims are not accepted, and the tested high-viscosity bulk-fill composites are not recommended to be placed in a single increment restoration, which is 4 mm thick.

CONCLUSIONS

Following 1-hour post-cure, the tested composites showed insufficient DC and low surface hardness values at 2-4 mm depth when irradiated for 20 and 40 seconds. The extension of curing time to 40 seconds improved the DC and MH values. MH was positively affected by increasing the amount of DC. Tetric N-Ceram showed the highest DC values, whereas Beautifil-Bulk displayed the highest MH values. Moreover, Opus Bulk Fill presented the lowest DC and MH values.

CONFLICT OF INTEREST

The authors declare no potential conflict of interests with respect to the authorship and/or publication of this article.

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