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ORIGINAL ARTICLE

Monomer conversion and shrinkage force kinetics of low-viscosity bulk-fill resin composites

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Abstract
Objective. To investigate the subsurface degree of conversion (DC) and shrinkage force formation of low-viscosity (flowable) bulk-fill composite materials. ***Materials and methods.*** Three flowable bulk-fill resin composites [SureFil SDR flow (SDR; Dentsply DeTrey), Venus Bulk Fill (VB; Heraeus Kulzer) and x-tra base (XB; VOCO)] and one conventional flowable control composite material [EsthetX flow (EX; Dentsply DeTrey)] were tested. The materials were photoactivated for 20 s at an irradiance of 1170 mW/cm^2 and the DC ($n = 5$) was recorded at 0.1-, 1.5- and 4-mm depth using Fourier transform infrared spectroscopy. Shrinkage forces ($n = 5$) of 1.5-mm-thick specimens were continuously recorded for 15 min using a custom-made stress analyzer. Data were statistically analyzed by ANOVA, Tukey's HSD and Bonferroni's post-hoc tests ($\alpha = 0.05$). ***Results.*** SDR generated the significantly lowest shrinkage forces (22.9 ± 1.4 N), but also attained the significantly lowest DC at 1.5-mm depth (67.5 ± 0.8%). The conventional flowable composite EX generated the significantly highest shrinkage forces (40.7 ± 0.7 N) and reached a significantly higher DC (74.4 ± 1.3%) compared to SDR and XB at 1.5-mm depth. The shrinkage force values of VB (29.4 ± 1.1 N) and XB (28.3 ± 0.6 N) were similar ($p > 0.05$). All materials attained significantly higher DC at 4-mm depth than at the near-surface. ***Conclusion.*** The tested low-viscosity bulk-fill materials show lower shrinkage force formation than a conventional flowable resin composite at high levels of degree of conversion up to 4-mm incremental thickness.

Key Words: *Bulk-fill materials, flowable resin composites, polymerization, shrinkage stress*

Introduction

Bulk-fill resin composites are an innovative class of dental composite materials, developed to simplify and shorten the placement of direct composite restorations [1,2]. They are fundamentally different from all other groups of resin composites by the fact that, according to manufacturers' claims, they can be efficiently photopolymerized at depths up to 4–5 mm and maintain low polymerization shrinkage stress at the same time.

While a high degree of monomer-to-polymer conversion is important to ensure material biocompatibility and good mechanical properties [3–7], it commonly entails high volumetric shrinkage in resin-based composite materials [8]. There is no proven correlation between the dimensional change of dental composite restorations and their clinical outcome [9]. However, polymerization shrinkage is highly correlated to polymerization stress, which is known as a contributing factor for restoration failure [9]. This polymerization-induced stress, being the result of confining the shrinking material by bonding to cavity walls, is considered responsible for a series of clinical complications including tooth-composite interfacial debonding [10], enamel cracking [11], cuspal deflection [12], post-operative sensitivity [13], and secondary caries [14]. Besides volumetric shrinkage and imposed cavity restrictions, the visco-elastic behavior of the material, usually described in terms of elastic modulus development and flow capacity, is an important determinant in the development of polymerization stress [15]. Before

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the onset of gelation, viscous flow compensates for most of the polymerization contraction due to polymer re-arrangement, causing stress relaxation. For this reason, shrinkage stress is reduced to much lower values than the expected theoretical values, which are calculated from the material's elastic modulus and shrinkage strain [16,17].

The degree of conversion has a fundamental influence on shrinkage stress, due to its inherent connection to the development of polymerization contraction and elastic modulus [15,18–20]. During the early phase of the polymerization reaction, shrinkage stress increases gradually in an almost linear manner with conversion [18]. As the vitrification stage is approached, however, stresses rise exponentially, with small increments in conversion leading to significant stress increase, due to the high stiffness of the cross-linked polymer network [18]. The necessity to include degree of conversion measurements in studies investigating the shrinkage stress of resin composites has been emphasized [15,18]. However, in the assessment of the material's behavior, it is mandatory to take into consideration also other factors that determine the ultimate shrinkage stress level, such as system compliance and cavity configuration [18,21].

Bulk-fill resin composites of low viscosity require the placement of a final capping layer made of a regular composite when used in direct restorations [22]. Their depth of cure [23–26], degree of conversion [26–28] and polymerization shrinkage [29] have been documented, as well as their mechanical properties [26,28,30]. However, information on the clinically relevant shrinkage stress of low-viscosity bulk-fill resin composites is scarce [31]. Manufacturers' claims regarding 4-mm depth of cure have been confirmed for the majority of bulk-fill composite materials [24–26] and even increased curing depths up to 8 mm have been described for some materials, but with a low degree of conversion and significantly reduced biaxial flexure strength and Vickers hardness [32]. It has been established that flowable bulk-fill materials can reach a clinically acceptable degree of conversion, although some of them are on the lower limit of ~55% monomer-to-polymer conversion [27].

The polymerization process of various bulk-fill composite materials has been identified to occur at a slower rate when compared with conventional resin composites [31,33]. Reducing the rate of polymerization has been shown to be beneficial in terms of reduced shrinkage stress formation due to the increased opportunity for viscous flow and polymer chain relaxation before mobility is restricted by vitrification [19,34]. On the other hand, any stress reduction achieved at the expense of adequate monomer conversion is clinically not intended, because a low degree of conversion might compromise both the mechanical and biological properties of the restoration [3–7].

The aim of this study was to investigate the shrinkage force kinetics and subsurface monomer conversion of flowable bulk-fill resin composites. The tested null hypotheses were that flowable bulk-fill materials and a conventional flowable resin composite would not differ in: (i) shrinkage force formation and (ii) the degree of conversion attained at the near-surface (0.1 mm) and at 1.5- and 4-mm depth.

Materials and methods

Three flowable bulk-fill resin composites [SureFil SDR flow (Dentsply DeTrey, Konstanz, Germany), Venus Bulk Fill (Heraeus Kulzer, Hanau, Germany) and x-tra base (VOCO, Cuxhaven, Germany)] and one conventional microhybrid flowable control material [EsthetX flow (Dentsply DeTrey)] were used in this study. The composition of the materials is presented in Table I. In all subsequent tests, the composite specimens were photoactivated for 20 s with a LED light-curing unit (Bluephase G2; Ivoclar Vivadent, Schaan, Liechtenstein) in high-intensity mode. The output irradiance (1170 mW/cm^2) was measured using a calibrated USB 4000 spectroradiometer (Ocean Optics, Dunedin, FL) and verified periodically during the experiments.

Degree of conversion

Composite specimens (diameter: 10 mm, thickness: 0.1 mm) were prepared by compressing uncured

Table I. Manufacturers' information about the tested composite materials.

Composite material	Manufacturer	Shade/ LOT	Resin composition	Filler amount (wt%/vol%)
SureFil SDR flow	Dentsply DeTrey, Konstanz, Germany	Universal/ 120228	Modified UDMA, Bis-EMA, TEGDMA	68/45
Venus Bulk Fill	Heraeus Kulzer, Hanau, Germany	Universal/ 010031	Bis-EMA, UDMA	65/38
x-tra base	VOCO, Cuxhaven, Germany	Universal/ 1144252	Bis-EMA, UDMA	75/60
EsthetX flow	Dentsply DeTrey, Konstanz, Germany	A2/ 110617	Bis-GMA adduct, Bis-EMA, TEGDMA	61/53

wt%, weight percentage; vol%, volume percentage; UDMA, urethane dimethacrylate; Bis-EMA, ethoxylated bisphenol-A-dimethacrylate; TEGDMA, triethylene glycol dimethacrylate; Bis-GMA, bisphenol-A-glycidyldimethacrylate.

composite paste between two Mylar strips under 10^7 Pa (Carver press; Specac Ltd., Orpington, Kent, UK). For near-surface measurements (0.1 mm), the specimens were irradiated with the light guide tip of the curing unit in direct contact to the upper Mylar strip using white paper as a background. For measurements at a particular depth, uncured overlays (diameter: 10 mm) of the respective composite material were placed in appropriate thickness (1.5 or 4 mm) above the upper Mylar strip and irradiation was performed in direct contact of the light guide tip to a Mylar strip covering the top of the overlay. The degree of conversion ($n = 5$ per group) was measured 15 min after irradiation using Fourier transform infrared spectroscopy (Model 2000; Perkin Elmer, Beaconsfield, Bucks, UK) [35]. Recording and processing of absorption spectra of the composite specimens were carried out with Spectrum v5.3.1 software (Perkin Elmer). Spectra of unpolymerized and polymerized composite specimens were recorded in transmission mode at room temperature (22°C), corrected by subtracting the background and then converted into the absorbance mode. The spectra of unpolymerized specimens were taken after pressing the specimens into pellets of spectroscopically pure potassium bromide (Merck, Darmstadt, Germany). A total of 20 scans per specimen were measured at a resolution of 4 cm^{-1}. The peak ratios were calculated according to Rueggeberg et al.'s [36] baseline method. For all materials except SureFil SDR flow, the degree of conversion (DC) was calculated from the equivalent aliphatic (1638 cm^{-1})/aromatic (1610 cm^{-1}) molar ratios of cured (C) and uncured (U) specimens according to the following equation:

$$DC (\%) = (1 - C/U) \times 100$$

In case of SureFil SDR flow, the peak at 1600 cm^{-1} was used as an internal reference due to the absence of the aromatic carbon bond [26].

Shrinkage force measurements

Measurements of polymerization shrinkage force were carried out using a custom-made stress analyzer (Figure 1), described in detail elsewhere [37,38]. In brief, the upper part of the device consisted of a semi-rigid load cell (PM 11-K; Mettler, Greifensee, Switzerland; instrument compliance: 0.4 μm/N), to which a metal cylinder was screwed. A standardized volume of composite material (42 mm^3) was applied to the front edge of the cylinder. The material was compressed to a thickness of 1.5 mm and a surface area of 28 mm^2 at the top and at the bottom of the specimen (corresponding to a ratio of bonded to unbonded surface area, i.e. C-factor, of 2.0), by means of a glass plate attached to the base of the device. To improve adhesion, the surfaces of the metal cylinder and of the glass plate were sandblasted with 50-μm Al_2O_3 and primed or silanized (Monobond Plus; Ivoclar Vivadent). Photoactivation was performed through the glass plate, via a recess in the lower frame, at a standardized distance of 1 mm from the test material. The forces generated during polymerization shrinkage were detected by means of the load cell at a sampling frequency of 5 Hz and continuously recorded over a period of 15 min from the initiation of photoactivation. Data were transferred in real-time to an attached computer (Macintosh IIfx; Apple Computer, Cupertino, CA) by means of an A/D converter using custom-made software. Five specimens were tested per experimental group.

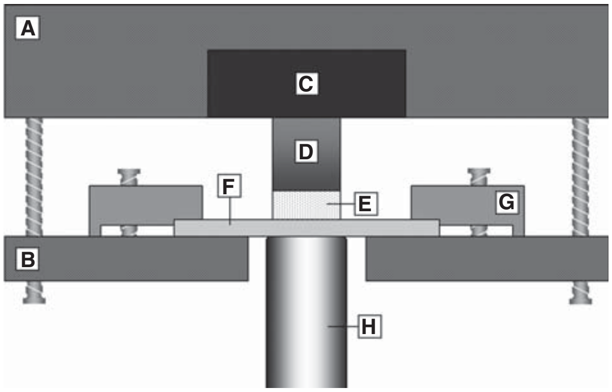

Figure 1. Diagram of the measuring device for shrinkage force (reproduced with permission from an earlier publication [37]). (A) Upper part of measuring device; (B) Lower part of measuring device; (C) Load cell; (D) Metal cylinder; (E) Composite specimen; (F) Glass plate; (G) Holder of glass plate; (H) Curing light tip.

Statistical analysis

After confirming the validity of the assumption of normality by means of the Kolmogorov-Smirnov and Shapiro-Wilk tests, the degree of conversion and shrinkage force data were analyzed using one-way analysis of variance (ANOVA). Bonferroni's post-hoc test was used for pairwise comparison of the degree of conversion between different depths (within the same material) and between different materials (within the same depth), respectively, whereas Tukey's HSD post-hoc test was used to detect pairwise differences in shrinkage force data. Two-way ANOVA and partial eta-squared statistics were applied to investigate the influence of the parameters 'composite material' and 'measuring depth' on the degree of conversion. The level of significance was set at 5%. All statistical analyses were performed using SPSS 20.0 software (SPSS Inc., Chicago, IL).

Results

Table II summarizes the results of the degree of conversion and shrinkage force measurements of the tested materials. Two-way ANOVA revealed significant differences in the degree of conversion due to both 'composite material' ($p < 0.001$) and 'measuring

Table II. Mean degree of conversion (%) of the tested composite materials at 0.1-, 1.5- and 4-mm depth, and mean shrinkage force (N) measured 15 min after photoactivation. Different uppercase letters indicate significant differences between materials at the same depth, whereas different lowercase letters indicate significant differences within the same material, but at different depths ($p < 0.05$).

		SureFil SDR flow	Venus Bulk Fill	x-tra base	EsthetX flow
Degree of conversion (%)	0.1 mm	61.4 (1.4) ABa	66.8 (0.4) Ca	59.2 (0.8) Aa	62.0 (1.8) Ba
	1.5 mm	67.5 (0.8) Ab	72.9 (1.3) Cb	69.6 (0.7) Bb	74.4 (1.3) Cc
	4 mm	70.0 (0.9) Bc	78.8 (1.1) Cc	67.5 (0.8) Ab	66.1 (1.3) Ab
Shrinkage force (N)		22.9 (1.4) A	29.4 (1.1) B	28.3 (0.6) B	40.7 (0.7) C

Standard deviations are given in parentheses.

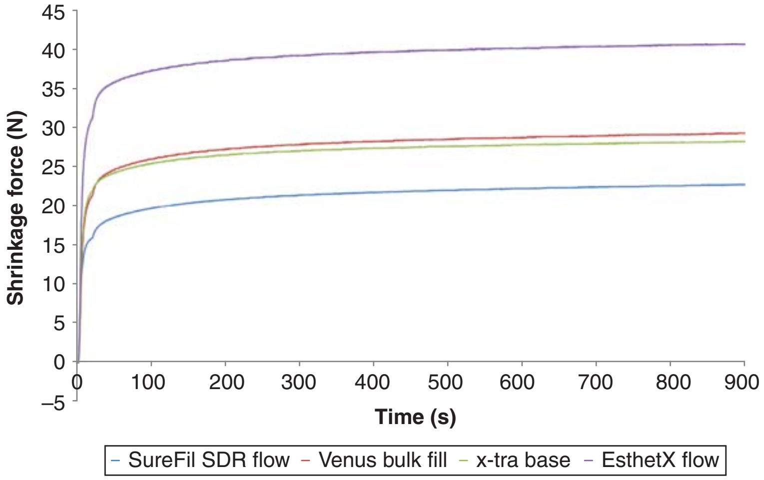

Figure 2. Mean polymerization shrinkage force curves of the tested materials as a function of time.

depth' ($p < 0.001$), and a significant interaction effect was found between these two factors ($p < 0.001$). The 'measuring depth' ($\eta^2 = 0.943$) had a stronger influence on the degree of conversion than the 'composite material' ($\eta^2 = 0.896$).

Venus Bulk Fill was the significantly highest polymerized material at 0.1- and 4-mm depth, but at 1.5-mm depth, the conventional flowable resin composite EsthetX flow reached a similar degree of conversion ($p = 0.258$). At the near-surface (0.1 mm), x-tra base attained the lowest degree of conversion, followed by SureFil SDR flow in the same statistical group, while at 1.5-mm depth, SureFil SDR flow achieved the significantly lowest degree of conversion. At 4-mm depth, the conventional flowable control material EsthetX flow reached the lowest degree of conversion, which was, however, not significantly different from the degree of conversion attained by x-tra base. All materials exhibited significantly higher degrees of conversion at 1.5-mm depth than at the near-surface. Furthermore, at 4-mm depth, SureFil SDR flow and Venus Bulk Fill even reached significantly higher degrees of conversion than at 1.5-mm depth, while the conventional flowable composite showed a significant decrease in the degree of conversion from 1.5- to 4-mm depth.

Figure 2 shows the time-dependent development of shrinkage force for each tested material. One-way ANOVA revealed significant differences in shrinkage force between the materials ($p < 0.001$). The conventional flowable resin composite EsthetX flow generated the significantly highest shrinkage forces (40.7 ± 0.7 N). The significantly lowest shrinkage forces were generated by SureFil SDR flow (22.9 ± 1.4 N), while Venus Bulk Fill (29.4 ± 1.1 N) and x-tra base (28.3 ± 0.6 N) created intermediate shrinkage forces, not being significantly different from each other.

Discussion

Bulk-fill resin composites present an interesting alternative to conventional composite materials due to their simplified use and reduced time needed for direct adhesive restorations. The present study evaluated clinically relevant parameters: the polymerization-induced shrinkage forces and the degree of conversion of low-viscosity bulk-fill resin composites. It established the reduction of shrinkage force formation for flowable bulk-fill materials compared to a conventional flowable resin composite, at high levels of degree of conversion.

Shrinkage stress is a local physical condition, not a basic property, and, as such, stress values vary depending on the testing system used, due to differences in geometries, test configurations and system compliance [39,40]. The most frequent method of measuring shrinkage stress is the tensilometer [41–43], where force generation of a composite material being bonded to two opposing surfaces is recorded by a strain-gauge load cell. In such a test set-up, only the forces developing uniaxially, in the long axis of the specimen, are registered, even though the shrinking material develops a triaxial stress state, as revealed by finite-element analysis [44]. The values registered by the load cell are influenced by the elongation that parts of the testing assembly may present when subjected to shrinkage force (i.e. compliance) and an inverse relationship between compliance and shrinkage stress has been described [45,46]. Near-zero compliance test set-ups, containing feedback systems in order to maintain the original height of the specimen throughout the experiment, might overestimate stresses associated with the clinical situation, where deformation of dental substrates would relieve part of the shrinkage force [15,47]. In the experimental set-up of the current investigation, axial specimen deformation was only partially restricted given that the load cell was axially displaced by 0.4 μm/N, resulting in a maximal deformation of 17 μm. In this way, a semi-rigid configuration of a cavity with a C-factor of 2.0 was simulated. Several studies have revealed that the cusps of premolars and molars deflect inwards after the placement of Class II resin composite restorations, with the amount of intercuspal narrowing ranging from 11–46 μm [48–50], thus justifying the experimental set-up in the present research. Furthermore, stress data obtained using a high compliance testing system showed a stronger correlation with microleakage and marginal gap formation than data from a low (near-zero) compliance system, besides a significant correlation with bond strength values [51].

SureFil SDR flow generated the significantly lowest shrinkage forces of all materials evaluated, which might be at least partially related to the lowest degree of conversion attained at 1.5-mm depth and, thus, at the bottom surface of the shrinkage force specimens. The degree of conversion is an important factor known to affect polymerization stress development through its influence on volumetric shrinkage [52]. In materials with high degrees of conversion, stress is increased due to restricted stress-relieving viscous flow and reduced molecular mobility in highly cross-linked polymer networks [39]. The peculiarity of SureFil SDR flow is the composition of its resin matrix, having a so-called 'polymerization modulator' incorporated in the high-molecular-weight urethane dimethacrylate resin backbone of the material [33]. The modulator is supposed to increase monomer flexibility and, thus, contribute to polymer matrix relaxation [33], evidently leading to lower shrinkage force formation [37]. Furthermore, the high molecular weight of the monomer is responsible for low polymerization shrinkage of SureFil SDR flow [37]. Finally, the polymerization process of SureFil SDR flow has been shown to occur at a slower rate when compared with conventional resin composites, thus delaying gelation, which allows more viscous flow of the material towards bonded surfaces and, thus, relieves part of the polymerization-induced shrinkage forces [33].

Venus Bulk Fill and x-tra base also showed significantly reduced shrinkage forces compared with the conventional flowable control material EsthetX flow. Even though the shrinkage force values of the two bulk-fill materials were similar, the backgrounds for that behavior are different. x-tra base represents the highest filled resin composite of the tested materials with a filler content of 60 vol%. Increased filler content has been associated with lower volumetric shrinkage, due to the fact that the volume occupied by organic matrix and, therefore, the number of reactive methacrylate groups decreases [53]. Lower total polymerization shrinkage might be responsible for the reduced shrinkage forces generated by x-tra base in comparison with the conventional flowable composite. Indeed, a direct relationship between volumetric shrinkage and polymerization stress has been established in semi-rigid testing systems [54].

Contrary to x-tra base, Venus Bulk Fill possesses the highest resin content among the materials evaluated in the current investigation. Despite the overall highest degree of conversion of Venus Bulk Fill, the material also generated lower shrinkage forces than the conventional flowable composite EsthetX flow. A recent study revealed that Venus Bulk Fill exhibited a low maximum stress rate and the longest time to achieve this maximum stress rate when compared to other high- and low-viscosity bulk-fill materials [31]. Similarly to SureFil SDR flow, a prolonged pre-gel phase probably allowed more shrinkage stress relief by viscous flow. According to all stated above, the first null hypothesis was rejected, given that all bulk-fill materials generated significantly lower shrinkage forces than the conventional flowable control composite.

To achieve up to 4-mm depth of cure, manufacturers of flowable bulk-fill materials took advantage of the fact that a relatively low filler amount (Venus Bulk Fill) or enlarged filler size (SureFil SDR flow, x-tra base) increases the translucency of the composite material [30]. Light propagation is then enhanced due to the reduced filler-matrix interface area, which decreases light scattering. This might have led to the observed high levels of degree of conversion throughout the depth of the bulk-fill materials evaluated in the current investigation. Venus Bulk Fill exhibited the highest degree of conversion among the tested bulk-fill resin composites at 4-mm depth, followed by

SureFil SDR flow and x-tra base, which is in good agreement with the sequence presented in other papers [26,27,32]. The unexpected high degree of conversion of the conventional flowable control material EsthetX flow up to 4-mm depth might also be ascribed to its high translucency. The fact that the control material was used in shade A2, and thus in a light shade, probably contributed to this observation. It is well known that light composite shades have higher transmission coefficients and allow deeper light penetration than darker shades, which results in higher curing depths [55,56]. In addition to that, EsthetX flow contains triethylene glycol dimethacrylate (TEGDMA), a diluent monomer of low molecular mass, well known for its capacity to increase monomer conversion [57]. Due to the high degree of conversion attained by the conventional flowable control material, the second null hypothesis could only be partially rejected. Even at 4-mm depth, the conventional flowable resin composite reached a similar degree of conversion as one of the bulk-fill materials under investigation (x-tra base).

An interesting phenomenon observed in the present study was the increase in the degree of conversion of all materials at 1.5-mm depth when compared to the near-surface values (0.1 mm). The same effect has been described by Czasch and Ilie [26] for Vickers hardness and indentation modulus values of flowable bulk-fill materials. In addition, Asmussen and Peutzfeldt [58] noticed increased hardening of conventional resin composites in deeper parts of the material than in the more superficial parts. A plausible explanation might be that heat formation due to the exothermic nature of free radical bulk polymerization gives rise to an increase in the degree of conversion in deeper parts of a bulky specimen [58,59]. This is especially true for composite formulations with lower filler content, since the presence of fillers partially reduces the temperature increase [59]. Indeed, SureFil SDR flow and Venus Bulk Fill with lower filler contents than x-tra base and EsthetX flow even showed a continuing increase in degree of conversion up to 4-mm depth.

Conclusion

Under the conditions of the present *in vitro* study, the tested low-viscosity bulk-fill materials generated lower shrinkage forces compared to a conventional flowable composite at high levels of degree of conversion up to 4-mm depth. This may support the intended application of these materials in up to 4-mm layer thickness for restoring high C-factor and deep posterior cavities.

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