Correlation between Polymerization Shrinkage Stress and C-factor Depends upon Cavity Compliance

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Abstract

Objectives. The literature reports inconsistent results regarding using configuration factor (C-factor) as an indicator to reflect the generation of polymerization shrinkage stress (PS) from dental restorative composites due to the constraint of cavity configuration. The current study aimed at unraveling the complex effects of C-factor on PS based on analytical and experimental approaches together, such that the reported inconsistency can be explained and a significance of C-factor in clinic can be comprehensively provided.

Methods. Analytical models based on linear elasticity were established to predict PS measured in instruments (testing systems) with different compliances, and complex effects of C-factor on PS were derived. The analyses were validated by experiments using a cantilever beam-based instrument and systematic variation of instrumental compliance.

Results. For a general trend, PS decreased with increasing C-factor when measured by instruments with high compliance. However, this trend gradually diminished and eventually reversed (PS became increased with increasing C-factor) by decreasing the system compliance.

Significance. Our study indicates that the correlation between PS and C-factor are highly dependent on the compliance of testing instrument for PS measurement. This suggests that the current concept on the role of C-factor in the stress development and transmission to tooth structures, higher C-factor produces higher PS due to reduced flow capacity of more confined materials, can be misleading. Thus, the compliance of the prepared tooth (cavity) structure should also be considered in the effect of C-factor on PS.

Keywords: Dental restorative materials; Photopolymerization; Polymerization shrinkage stress; C-factor; Compliance of constraint

Introduction

In dental restorations, photo-polymerized composites as restorative materials are confined by the cavity walls, result of which could cause considerable polymerization shrinkage stress (PS) developed at the restorative-tooth interface during and after polymerization that can impart interfacial/cohesive failures and tooth decay from subsequent bacterial biofilm ingrowth [1-3]. Feilzer *et al.* developed the concept of configuration factor (C-factor) for the relationship between the confinement (cavity configuration) and stress development [4]. The C-factor defines the relationship between the free and restrained composite surface area of a dental restoration and is used to correlate with the risk of PS-induced restorative failure in clinic. In order to correlate the measured value of PS to the degree of confinement in experiments, C-factor has been calculated as the ratio of bonded surface area to unbonded area of a sample in the test [4]. However, studies in evaluating polymerization stress have been showing inconsistent results on the correlation of PS and C-factor and thus, using C-factor as an index for the risk analysis has long been controversial. The objective of this study is to systematically investigate complex effects of C-factor on PS development, such that the inconsistent results reported can be

explained and a comprehensive significance of C-factor for clinic-relevant environment can be provided.

Inconsistent results of the C-factor effect on PS appear in the studies using testing instruments with different compliances. Most experimental evaluations of the aforementioned PS have been using a force transducer mechanism to record the uniaxial force induced from a disk/cylinder-shape specimen under axial constraints [5]. Although the basic principle of those experiments is the same, differences in the instrumental compliance exist among them. The instruments reported in the literature vary from an ideally rigid one (zero compliance) to a considerably compliant one [6, 7]. In order to obtain the zero-compliance, the sample height is maintained constant during the polymerization shrinkage by a servo-controlled feedback unit of the instrument. Using this type of instrument, the higher the C-factor (*i.e.* the higher the ratio of the bonded surfaces) the higher the PS was observed [4, 8-11]. This trend was intuitively explained by that, as the C-factor increases (*i.e.* the ratio of free surfaces decreases), the capacity of material deformation (flow) and consequent stress relief decreases. Accordingly, a low C-factor was advised to be maintained in clinic (*e.g.* using incremental filling technique instead of bulk filling) to facilitate the stress relief [4, 8]. Quite a few studies (either *in vitro* or *ex vivo*) in the literature [12-16] evaluated effects of C-factor on marginal adaptation of restorations, and the results indeed showed that cavity with lower C-factor was associated with a lower chance of interfacial failure such as microleakage and marginal gap. Thus, C-factor has been acknowledged as a valid variable in experiments to describe the development of PS due to constraints. In instruments with certain compliance (*e.g.* cantilever beam-based instruments) [7, 17, 18], which provide a more clinically-relevant testing environment as compliant tooth walls, force generated by the force transducer for balancing the polymerization shrinkage is registered as the PS. Using

such compliant instruments, some studies indicated the higher the C-factor the lower the PS (opposite to the findings using ‘zero compliance’ instruments) [7, 17, 19-22], while the other studies presented no obvious dependency between PS and C-factor [10, 23]. Thus, the validity of C-factor as a PS predictor has been questioned.

In this study, combined analytical and experimental approaches were adopted to investigate the complex correlations between C-factor and PS development under various instrumental compliances. The role of sample volume in the C-factor effect on PS development is also discussed since samples can have same C-factor value but different volumes and *vice versa*. Analytically, a simplified model based on the linear elasticity was established to predict PS of restorative materials tested under rigid and compliant systems. Thus, effects of C-factor and sample volume on PS development were derived mathematically. The analytical results were validated by experiments using a NIST-developed cantilever beam-based instrument [24, 25]. The effect of instrument compliance on the measurement of polymerization stress was examined by changing the position of restorative composite sample along the length of the cantilever beam, such that the variable instrument compliance can mimic the compliance of the prepared tooth cavity. Our results show that the correlation between PS and C-factor is highly compliance-dependent. For a general trend, PS decreased with increasing C-factor when measured by instruments with high compliance. However, this trend gradually diminished and eventually reversed (PS became increased with increasing C-factor) by decreasing the system compliance. This compliance-dependent effect of C-factor on PS not only explains the numerous inconsistent results reported in the literature; it also provides new insights into the concept of C-factor for clinic-relevant environment and useful guidelines for clinical selection of filling technique in order for a reduced PS.

Experiments[♦]

In this investigation, a commercial dental composite (TPH SpectraTM, Dentsply-Caulk, Milford, DE) was used as the testing material. TPH is a blue light activated, urethane modified Bis-GMA/TEGDMA (50:50 mass ratio) based composite, filled primarily with barium boron aluminum silicate glass particles at 78 wt.% (≈ 57 vol.%).

The effects of C-factor on polymerization stress (PS) development were examined using a NIST-developed cantilever beam-based instrument (tensometer) [24, 25]. The instrument was constructed according to design criteria following first principles of mechanics to provide the accuracy and sensitivity of the PS measurement. The desired compliance of the testing instrument can be easily achieved by varying the sample position along the length of the cantilever beam or by using a beam with different height or different material. In this study, an instrumental compliance (C) ranging from 0.33 to 2186.06 $\mu\text{m}/\text{N}$ was obtained (Supplementary Data (SD), Fig. S1 and Table S1). Using this instrument (see Fig. 1a and 1b), an uncured specimen in disk shape was bonded to two flat quartz rods that had been treated with a methacrylate silane to promote adhesion between the specimen and rods. The lower rod was fixed and the upper rod attached to the cantilever beam. A non-tacky teflon sleeve with an injection hole and a smaller air-venting hole was used to encase the rods and the specimen. As curing light ($500 \pm 10 \text{ mW}/\text{cm}^2$ intensity and 40 s irradiation through a 10 mm-diameter light guide for all the tests, QHL75TM curing lamp, Dentsply-Caulk, Milford, DE) transmitted through the lower rod into the specimen, polymerization shrinkage occurred and the resulting stress

[♦] Certain commercial materials and equipment are identified in this manuscript in order to specify adequately the experimental and analysis procedures. In no case does such identification imply recommendation or endorsement by the National Institute of Standards and Technology (NIST) nor does it imply that they are necessarily the best available for the purpose.

caused a deflection in the beam. This deflection was recorded by a displacement sensor at the free end of the beam and used to calculate the PS based on a beam formula as shown in Fig. 1c. More details on the engineering mechanics and design of the instrument can be found in our previous publications [24, 25].

Various C-factors for the specimen were achieved by adjusting either its diameter or height (by changing the diameter of quartz rods or the gap between the rods). For a specimen of diameter $2r$ and height L_s , the C-factor (C_f), defined as the ratio of bonded surface area to unbonded area of sample, is:

$$C_f = \frac{2\pi r^2}{2\pi r L_s} = \frac{r}{L_s} \quad (1)$$

The sample configurations (r and L_s) tested in this study, the C-factor (C_f) and the sample volume (V) in each case are given in the SD, Table S2. For each set of the experiments (*i.e.* each C-factor and each instrumental compliance tested), PS development was monitored continuously for 30 min with 10 Hz sampling temporal resolution and at least three replicates were conducted. A typical curve for the real-time development of PS is shown in Fig. 1d. All experiments and sample preparations were performed at room temperature and under a yellow light environment avoiding premature photopolymerization.

Results and Discussion

Most of known instruments for the polymerization stress (PS) measurement are based on the use of a force transducer (load cell) to perceive forces through its deformation. The essential difference among these instruments is the compliance included ([see detailed analyses in \[26\]](#))

and we divide them into two categories of mechanism: displacement-controlled and force-controlled.

The displacement-controlled instruments are commonly-recognized as ‘rigid’ or ‘zero-compliance’ instruments, in which any sample shrinkage sensed by the load cell is immediately compensated for by a full feedback response exerted by a servo-controlled unit incorporated in the instrument [4, 11]. Thus, the final position of sample is fixed and the force registered for the compensation is used as PS. For a uniaxially-constrained sample (*e.g.* in this study), the shrinkage and the full feedback response, that occur continuously during the process can be described as three separate steps illustrated in Fig. 2a: (1) free axial shrinkage (free of constraint), (2) sample elongation, δ_1 , by the load cell, and (3) sample elongation, δ_2 , by the feedback unit until the initial sample height (L_s) is achieved. The combination of elongations in the second and third steps, denoted as δ_r shown in Fig. 2a, is considered as a whole feedback response associated with PS. Accordingly, assuming elastic responses, the PS in the displacement-controlled system, σ_r , can be theoretically approximated as:

$$\sigma_r = E\varepsilon_r = \frac{E\delta_r}{L_f} = \frac{EL_s\varepsilon_f}{L_s(1-\varepsilon_f)} = \frac{E\varepsilon_f}{1-\varepsilon_f} \quad (2)$$

where E is the elastic modulus of the sample, ε_r is the elastic strain exerted by the displacement-controlled feedback, $\varepsilon_f = \frac{L_s - L_f}{L_s}$ is the free shrinkage, and L_f is the sample height corresponding to the free axial shrinkage.

The force-controlled instruments refer to those without using the aforementioned servo-controlled unit and are commonly-recognized as ‘compliant’ instruments (*i.e.* instrumental

compliance allowed). During the evolution of polymerization shrinkage, the stress measured by the load cell for balancing the sample uniaxial shrinkage is registered as PS. This balancing process can be decomposed into two steps shown in Fig. 2b: (1) free axial shrinkage, and (2) feedback response by the load cell. The cantilever beam-based instrument (tensometer) used in this study falls under the category of force-controlled instruments, and the polymerization stress (PS), σ_c , can be calculated as:

$$\sigma_c = \frac{k\Delta}{A} \quad (3)$$

where k is the spring constant (or stiffness) of the load cell (*i.e.* the cantilever beam for the tensometer instrument), which equals to the inverse of the instrumental compliance ($k = 1/C$); Δ is the effective deformation of the load cell; and A is the cross-sectional area of the sample. σ_c can be also derived as:

$$\sigma_c = E\varepsilon_c = \frac{E\delta_c}{L_f} = \frac{E\delta_c}{L_s(1-\varepsilon_f)} \quad (4)$$

where ε_c and δ_c are the elastic strain and axial elongation imposed by the force-controlled feedback, respectively. Given the relationship of $\delta_c + \Delta = L_s\varepsilon_f$ and combining Equations (3) and (4) one can obtain:

$$\sigma_c = \frac{kE\varepsilon_f}{k_s + k(1-\varepsilon_f)} \quad (5)$$

where $k_s = AE / L_s$ is the spring constant of sample.

Stresses measured for samples with different C-factors are compared for the displacement-controlled and force-controlled instruments. Based on Equations (2) and (5), giving two samples with different C-factors, C_{f1} and C_{f2} and $C_{f1} > C_{f2}$, the corresponding PS obtained from the displacement-controlled system (σ_{r1} and σ_{r2}) can be compared as:

$$\sigma_{r1} - \sigma_{r2} = \frac{E_1 \varepsilon_{f1}}{1 - \varepsilon_{f1}} - \frac{E_2 \varepsilon_{f2}}{1 - \varepsilon_{f2}} = M_r (\varepsilon_{f1} - \varepsilon_{f2}) \quad (6)$$

where $M_r = E / (1 - \varepsilon_{f1})(1 - \varepsilon_{f2})$ is a positive term, and $E_1 = E_2 \equiv E$ is assumed. The corresponding PS obtained from the force-controlled system (σ_{c1} and σ_{c2}) can be compared as:

$$\begin{aligned} \sigma_{c1} - \sigma_{c2} &= \frac{kE_1 \varepsilon_{f1}}{k_{s1} + k(1 - \varepsilon_{f1})} - \frac{kE_2 \varepsilon_{f2}}{k_{s2} + k(1 - \varepsilon_{f2})} \\ &= M_c (\varepsilon_{f1}(k_{s2} + k) - \varepsilon_{f2}(k_{s1} + k)) \end{aligned} \quad (7)$$

where $M_c = kE / (k_{s1} + k(1 - \varepsilon_{f1}))(k_{s2} + k(1 - \varepsilon_{f2}))$ is a positive term as well. In order to determine the signs of Equations (6) and (7), correlation between ε_f and C_f is needed. This correlation was determined experimentally (see Section S1 in the SD for details of the ε_f measurement). The results shown in Fig. 3 indicate that, within the range of C_f studied, ε_f increased almost linearly with increasing C_f regardless how C_f was created (*i.e.* by changing either sample radius r , or height L_s , or both simultaneously). A similar trend has also been reported in the literature [27-29] and has been explained by that, the volumetric shrinkage, which is normally distributed in three dimensions, is gradually converted into only the axial direction with the continuous increase of C_f (*i.e.* the shrinkage in the radial direction is increasingly restricted due to the increasing boundary effect from the bonded surfaces). As a result, the shrinkage measured in the axial direction, ε_f , gradually shifts from the linear shrinkage (*i.e.* 1/3 of

the volumetric shrinkage if an isotropic shrinkage is assumed) to the volumetric shrinkage as C_f increases. Based on this trend, giving $C_{f1} > C_{f2} > 0$, the following relationship exists:

$$\varepsilon_{f1} \geq \varepsilon_{f2} \quad (8)$$

The equal sign in Equation (8) is only valid if both C_{f1} and C_{f2} are large (corresponding to thin disk samples in experiment) such that both ε_{f1} and ε_{f2} reach the maximum, *i.e.* the volumetric shrinkage [28, 29].

From the results shown in Fig. 3 and the literature [28, 29], it is also found the slopes of the ε_f vs. C_f curves (before ε_f plateaus for the results shown in [28] and [29]) are all less than 1. Therefore, the following relationship between C_f and ε_f can also be drawn:

$$\frac{C_{f1}}{C_{f2}} > \frac{\varepsilon_{f1}}{\varepsilon_{f2}} \quad (9)$$

Combining Equations (6) and (8), one can obtain the following relationship:

$$\sigma_{r1} - \sigma_{r2} = M_r(\varepsilon_{f1} - \varepsilon_{f2}) \geq 0 \quad (10)$$

Equation (10) indicates that, in general, PS measured in a displacement-controlled system increases with the increase of C-factor, which is in line with the experimental results shown in the literature [4, 8-11]. The experimental result obtained by Feilzer *et al.* [4] did reflect the effect of C_f on PS development became minimal when C_f exceeded a certain value, which corresponds to the situation of equal sign in Equation (10).

For a force-controlled system, the correlation between σ_c and C_f , *i.e.* the sign of Equation (7), cannot be determined using the relationships shown in Equations (8) and (9) alone. For this

reason, Equation (7) needs to be further simplified. In the force-controlled system, in general, there are two extreme cases in terms of the spring constant of load cell, k , and sample, k_s :

Case 1: $k \gg k_s$, *i.e.* $C = 1/k \rightarrow 0$, the force-controlled instrument approaches to a displacement-controlled (zero-compliance) one. By neglecting k_s in Equations (7) and combining with Equation (8) one can obtain:

$$\sigma_{c1} - \sigma_{c2} \approx M_c k (\varepsilon_{f1} - \varepsilon_{f2}) \geq 0 \quad (11)$$

Thus, PS measured in a low-compliance force-controlled instrument also increases with the increase of C-factor.

For a typical disk-shape sample of dental composite used for PS measurements reported in the literature, *e.g.* $r = 6$ mm, $L_s = 1$ mm, and $E \approx 10$ GPa, the spring constant of sample $k_s \approx 280$ N/ μ m. By contrast, the force-controlled instruments reported in the literature have a spring constant, k ranging from 0.05 to 10 N/ μ m [7, 20, 24, 25, 30]. Therefore, most of the PS measurement using force-controlled instruments falls into the following case:

Case 2: $k \ll k_s$, such that k can be neglected compared to k_s and Equation (7) becomes:

$$\sigma_{c1} - \sigma_{c2} \approx M_c (\varepsilon_{f1} k_{s2} - \varepsilon_{f2} k_{s1}) = M_c E \left(\frac{\varepsilon_{f1} A_2}{L_{s2}} - \frac{\varepsilon_{f2} A_1}{L_{s1}} \right) = M_c E \pi (\varepsilon_{f1} C_{f2} r_2 - \varepsilon_{f2} C_{f1} r_1) \quad (12)$$

Given that $C_{f1} > C_{f2}$, and $r_1 = r_2$ or $r_1 > r_2$, Equation (12) becomes:

$$\sigma_{c1} - \sigma_{c2} < 0 \quad (13)$$

since $\varepsilon_{f1} C_{f2} < \varepsilon_{f2} C_{f1}$ according to Equation (9). Equation (13) indicates that, for a compliant force-controlled instrument, the measured PS decreases with increasing C-factor when C-factor

is created by changing either sample radius r only ($r_1 > r_2$) or height L_s only ($r_1 = r_2$). This argument is in agreement with the results reported in the literature [7, 17, 19-22] and further supported by the experiments performed in this study (Fig. 4). However, the results shown in Fig. 4 indicate that the inverse influence of C-factor on PS gradually diminished as the instrumental compliance decreased. One can expect that if the instrumental compliance decreased further (not performed in the present study), the PS would become increase with increasing C-factor. This corresponds to a transition from Case 2 to Case 1.

When C-factor is created by changing sample radius and height simultaneously, the above correlation between PS and C-factor is no longer valid since the sign of Equation (12) cannot be determined ($r_1 > r_2$ or $r_1 < r_2$ given $C_{f1} > C_{f2}$). This judgement is also supported by the results shown in the literature and the experiments performed in the present study. In the literature, a few studies [19, 22] based on force-controlled instruments created C-factor by varying both variables simultaneously, and the results indeed showed weaker correlations between PS and C-factor compared to those with only one variable being varied. For the present study, by combining all the C-factors shown in Fig. 4, Fig. 5 displays the measured PS as a function of combined C-factor (*i.e.* simultaneous variation of r and L_s). One can see that globally, PS still decreased with increasing C-factor for all the instrumental compliances tested; however, this trend was locally distorted for certain C-factors achieved by both varied r and L_s . Specifically, it can be seen that samples with same C-factor achieved by different combinations of r and L_s (indicated by asterisks in Fig. 5) could generate significantly different PS, suggesting that C-factor alone was not able to fully describe the effect of sample configuration on PS.

Since C-factor is only a ratio value of the sample surfaces and does not reflect any information about the absolute mass (or volume) of the sample, numbers of studies in the literature have pointed out sample volume should be considered together with C-factor in studying the sample configuration effect on PS [11, 19, 21, 22, 31, 32]. However, similar to C-factor, the correlation between PS and sample volume has been shown complex and highly related to both the measurement instrument and the way how sample configuration is created (*i.e.* by changing either the diameter or the height of sample or both) [11, 21]. Based on the results shown above, a comprehensive understanding regarding the effect of sample volume on PS is also achieved as detailed below. According to the calculations for the C-factor ($C_f = r/L_s$) and the volume ($V = \pi r^2 L_s$), one can see if L_s is fixed, a direct correlation exists between these two variables, *i.e.* if $V_1 > V_2$, then $C_{f1} > C_{f2}$; by contrast, if r is fixed, the inverse correlation exists, *i.e.* if $V_1 > V_2$, then $C_{f1} < C_{f2}$. Therefore, for sample configurations created by fixing r and varying L_s , the general trend of PS versus V follows the opposite trend of PS versus C_f (see Fig. 6a and 6b for experimental validation); while for those created by fixing L_s and varying r , the trend of PS versus V follows the same as that of PS versus C_f (Fig. 6c and 6d). For a simultaneous variation of r and L_s , the correlation between PS and V also cannot be determined (Fig. 6e and 6f). These trends, together with the derivations of the combinative effect of C_f and V on PS shown in the SD, Section S2 agreed well with the experimental results reported in the literature [4, 11, 17, 20-22], and indicated the consistencies between the effects of C-factor and sample volume on PS.

In the literature, almost all the studies regarding the effect of C-factor on PS focus only on the final magnitude of PS since the final PS is believed the most critical index related to the interfacial failure in clinic; however, information about the kinetics of PS development for different C-factors are also important because the kinetics tell how fast the PS being built-up in

association with the evolving material properties (*e.g.* shrinkage and modulus) during the polymerization process. Experimental results shown in Fig. 7 give the kinetics of the development of axial shrinkage and PS for selected C-factors and instrumental compliances. It can be seen from Fig. 7a and 7b that a higher C-factor, due to a more restricted shrinkage in the radial direction [28, 29], corresponded to a higher axial shrinkage over the entire polymerization process. For the development of PS (Fig. 7c and 7d), a higher C-factor corresponded to a higher dynamic PS during the first few seconds of polymerization (*i.e.* faster-developed PS) but a lower PS afterwards. This faster-developed PS while lower final PS for the higher C-factor sample is supported by the force-controlled model given in Equations (11) and (13) as detailed below. During the polymerization process, modulus of the sample (corresponding to the spring constant, k_s) gradually develops as the material transforms from monomeric to rubbery and then to glassy state. Therefore, for a certain instrument compliance (*i.e.* constant k), the relationship of $k \gg k_s$ should be valid during the early stage of polymerization, which corresponds to a higher PS for a higher C-factor according to Equation (11). With the evolution of k_s upon further polymerization (vitrification), the relative magnitude of k and k_s reverses (*i.e.* $k \ll k_s$) and consequently, the correlation between the dynamic PS and C-factor also reverses according to Equation (13). A recent analytical study based on a more comprehensive mathematical model also showed the relative compliance between the external constraint ($1/k$) and the composite sample ($1/k_s$) is an important parameter in determining the PS development [33]; however, effects of C-factor on PS development associated with the change of the relative compliance were not considered.

It is worthy to note that the PS models derived in this study (Equations 2 and 5) are based on purely elastic responses of the sample and do not take into account the incremental change of the sample modulus and shrinkage during the evolution of polymerization; therefore, quantitative

agreements between the models and experiments are not expected. However, this does not influence the overall trends of PS versus C-factor for the extreme cases (Equations 11-13) predicted in this study (see Fig. S3 in the SD for more information). One might also concern that as the C-factor and instrumental compliance change, the uniformity and degree of polymerization of the sample might also change and contribute to the different PS responses. According to the material manufacturer, the amount of irradiation adopted in this study was more than enough to cure a 2 mm thick sample (maximum tested in this study). Thus, a uniform curing for all the sample geometries tested can be reasonably assumed. Also, degree of conversion (DC) measured by *in-situ* near infrared (NIR) spectroscopy shown in Fig. 8 confirmed that DC of cured sample did not change significantly as the C-factor and instrumental compliance changed. Therefore, the different correlations between PS and C-factor achieved in this study are solely due to the changes of sample geometry and compliance of external constraint.

Clinically, the remaining tooth in dental restoration is not a completely rigid system. The compliance of tooth cavity varies depending on the cavity geometry and remaining wall thickness. For example, the compliance of the remaining cusps of a Class-II cavity (cavity size: width ≈ 3 mm, depth ≈ 2 mm, and wall thickness ≈ 1 mm) has been measured to be $\approx 3 \mu\text{m/N}$ [20]; while the mean compliance of typical Class-I cavities has been computed to be $\approx 0.4 \mu\text{m/N}$ [34]. Also, the compliance is not uniformly distributed within a certain cavity. The lower parts of the cavity are usually associated with thicker walls and have the compliance much lower than that in the upper parts. This non-uniform distribution of the cavity compliance (corresponding to a non-uniform distribution of the PS) explains the experimental observations that the restorative composites become debonded from the lower dentin part of the cavity while keep attached to the upper enamel walls reported in the literature [35, 36]. Therefore, the measured $3 \mu\text{m/N}$ of the

Class-II cusp compliance could be representative of a rough upper bound for the cavity compliance in clinic. In our experiments, for the instrumental compliance less than 3 $\mu\text{m}/\text{N}$, the correlation between PS and C-factor is in general contradicting to the widely-accepted concept of C-factor in dentistry, in which a higher C-factor is expected to generate higher PS (*i.e.* a higher risk of interfacial failure). Although in the experiments, PS is measured uniaxially and not fully representative of the tri-axial stress that typically occur in clinic, it has been claimed that the major clinical consequences of PS, *e.g.* cusp deflection, microleakage, bond failure, *etc.* are mainly affected by the axially orientated stresses at the bonded surfaces [21, 37]. Thus, the results presented in this study (*i.e.* compliance-dependent PS – C-factor correlation) not only explain the inconsistent trends of PS versus C-factor/sample volume obtained from *in vitro* measurements reported in the literature [8-11, 19-23], they also provide new insights into why using incremental filling technique (reduced C-factor compared to bulk filling) does not always abate the PS-induced effects compared to bulk filling that has been extensively studied [38-40]. According to the present study, incremental filling should be adopted only for such cavities with very low compliance (*e.g.* Class-I cavity with thick tooth walls) in order for a reduced PS.

Conclusion

We have provided a comprehensive study (analytically and experimentally) on the interrelationships between polymerization shrinkage stress (PS), C-factor, sample volume, and compliance of testing instrument for dental restorative composites. The study shows that C-factor, as a descriptor of local flow (stress relief) capacity of material, is more reliably correlated to the rate (not necessarily the magnitude) of PS development. The correlation between C-factor and magnitude of PS is highly dependent on the compliance of testing instrument and the way experimental samples are created for the C-factor. This compliance-dependent PS – C-factor

correlation can explain the numerous inconsistent results reported in the literature. The study demonstrates that correlating higher C-factor to higher PS can be misguided if without considering the compliance effect on this correlation. From the clinical perspective, this compliance effect is highly relevant to the prepared tooth structure for restoration. Thus, the selection of filling technique (*e.g.* bulk vs. incremental) for reducing PS in clinic should be made according to the compliance of the tooth cavity.

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Figure 1. Schematic diagram (a) and photograph (b) of the NIST-developed tensometer instrument for the measurement of polymerization stress development. Schematic of the cantilever beam configuration for polymerization stress measurement (c); the stress (σ) is calculated as: $\sigma = \frac{F}{A} = \frac{6\delta EI}{\pi r^2 a^2 (3l - a)}$, where A is the cross-sectional area of the sample; E and $I (= wh^3/12)$ are the Young's modulus and the moment of inertia of the beam, respectively. Typical curve of the measured polymerization stress (PS) development with the data shown for the first 100 s after photo-activation (d).

Figure 2. Schematics of the analytical model for PS derivation in rigid (a), and compliant (b) measurement system. The top and bottom surfaces of the sample are radially-confined.

Figure 3. Axial shrinkage (ϵ_f) measured at 30 min after irradiation as a function of C-factor (C_f). Error bars indicate the standard deviation of the measurement. Line connecting the data points is provided for visual assistance. Data values are given in the SD, Table S3.

Figure 4. Polymerization stress measured at 30 min after irradiation (σ_c) normalized by instrument spring constant (k) as a function of C-factor (C_f) for various instrumental compliances. C-factors are created by either fixing sample radius (r) while varying sample height (L_s) with $r = 1.25$ mm (a), and $r = 3$ mm (b); or fixing L_s while varying r with $L_s = 1$ mm (c), and $L_s = 2$ mm (d). Error bars indicate the standard deviation of the measurement. Lines connecting the data points are provided for visual assistance. Data values for the polymerization stress are given in the SD, Table S4.

Figure 5. Polymerization stress measured at 30 min after irradiation (σ_c) as a function of combined C-factor (C_f) under two selected instrumental compliances: $2.62 \mu\text{m/N}$ (a), and $12.12 \mu\text{m/N}$ (b). Error bars indicate the standard deviation of the measurement. Asterisks inside the plots indicate same C-factor created by different combinations of sample radius, r , and height, L_s .

Figure 6. Effect of sample volume (V) on polymerization stress measured at 30 min after irradiation (σ_c). σ_c normalized by beam spring constant (k) as a function of V under various instrumental compliances (a-d). Sample configurations are created by either fixing sample radius (r) while varying sample height (L_s) with $r = 1.25$ mm (a), and $r = 3$ mm (b); or fixing L_s while varying r with $L_s = 1$ mm (c), and $L_s = 2$ mm (d). σ_c as a function of V (simultaneous variation of r and L_s) under selected instrumental compliances: 2.62

$\mu\text{m/N}$ (e), and $12.12 \mu\text{m/N}$ (f). Error bars indicate the standard deviation of the measurement. Lines connecting the data points are provided for visual assistance.

Figure 7. Selected plots for the real-time development of axial shrinkage (ϵ_f) and polymerization stress (PS). ϵ_f development for different C-factors created by fixing sample radius ($r = 1.25 \text{ mm}$) while varying height (a), or fixing sample height ($L_s = 1 \text{ mm}$) while varying radius (b); (c) and (d) show the corresponding plots for PS development.

Figure 8. Degree of conversion (DC) measured at 30 min after irradiation as a function of C-factor under three selected instrumental compliances. DC was measured by near infrared (NIR) spectrometer integrated into the NIST tensometer as detailed in our previous study [25]. Error bars indicate the standard deviation of the measurement.