Published in final edited form as:

Dent Mater. 2013 November ; 29(11): . doi:10.1016/j.dental.2013.08.198.

Shrinkage stress development in dental composites—An analytical treatment

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Abstract

Objectives—The aim of this paper is to develop a comprehensive mathematical model for shrinkage stress development in dental composites that can account for the combined effect of material properties, specimen geometry and external constraints.

Methods—A viscoelastic model that includes the composite's elastic, creep and shrinkage strains, and their interaction with the sample's dimensions and the external constraint is developed. The model contains two dimensionless parameters. The first one represents the compliance of the external constraint relative to that of the composite sample, and the second controls the rate of shrinkage stress decay through creep. The resulting differential equation is solved for two special cases: zero compliance and zero creep. Predictions for shrinkage stress measurements are then made using the analytical solutions for instruments with different compliances, samples with different thicknesses and composites with different filler fractions.

Results—The model correctly predicts how shrinkage stress increases with time, its dependence on the interaction between the entire system's compliance and the material properties, and the effect of the filler fraction on its maximum value. Comparisons with reported shrinkage stress measurements have provided very good agreement between theory and experiments.

Significance—The results provided by the model can help to resolve most, if not all, of the seemingly conflicting experimental observations reported in the literature. They can also provide some useful guidelines for optimizing the mechanical performance of dental composite restorations. The compliance ratio, a new parameter derived from the model, represents a fuller description of the constraints of the system.

Keywords

Dental composites; Shrinkage stress; Compliance; Filler fraction; Mathematical model

1. Introduction

Numerous studies [1–8], mostly experimental, have been performed to assess the shrinkage stress generated by different dental composites during curing. The shrinkage stress-measuring instruments reported in these studies use either a uniaxial tensile [1–3] or cantilever bending [4] configuration. In the former, a feedback control system is sometimes used to maintain the thickness of the composite sample, which is usually in the shape of a disk. Thus, different instruments have different compliances, and many studies have been

conducted to understand the interactive effect of the material's mechanical properties (mainly elastic modulus and shrinkage strain) and the instrument's compliance on the maximum shrinkage stress value that can be achieved [1,3,6–8]. However, many of the reported results appear to contradict with each other. As a result, debate is still ongoing with regard to the relative importance of these factors to the generation of shrinkage stress. In particular, the presumed benefit of increasing the filler fraction of the composite as an attempt to reduce the shrinkage strain and, hopefully, the shrinkage stress has been called into question, with some experimental results showing the exact opposite [3,5].

In contrast to experimental work, there have been few theoretical treatments of the topic of shrinkage stress development in dental composites. Previous attempts [9–13] only provided numerical solutions and property measurements without fully considering the effect of the compliance of the surrounding constraint on the shrinkage stress. It is the aim of this paper to fill this important gap in our study of this class of restorative materials by providing explicit analytical solutions for the problem using a simple Maxwell viscoelastic model based on engineering mechanics. Such analytical solutions will allow the shrinkage stress kinetics and the effect of the material properties and geometrical constraints to be analyzed explicitly and comprehensively.

2. Methods

Consider the model shown in Fig. 1, in which the shrinkage of a composite of cross-sectional area A and thickness l is constrained by a spring of stiffness k representing the stress-measuring device (or the surrounding tooth tissues). A one-dimensional viscoelastic model is used to describe the mechanical behavior of the composite material [14]. This is justified because shrinkage stress measurement is most commonly made in a uniaxial (1-D) direction. In classical mechanics, a viscoelastic model consists of two constituent elements: an elastic spring and a viscous dashpot. The two elements can be arranged in series (the Maxwell model) or in parallel (the Kelvin-Voight model). In the former case, the elements have the same stress and their strains combine to give the total strain of the model. In the latter case, the opposite is true. Given that the composite material can undergo permanent deformation, the Maxwell model is more appropriate. However, we need to add a third element to account for polymerization shrinkage. Thus, the net strain (ε) of the composite at all times consists of the sum of three components: the elastic strain (ε_e), the creep strain (ε_c) and the linear shrinkage strain (ε_s). Incrementally over a short time interval dt ,

$$d\varepsilon = d\varepsilon_e + d\varepsilon_c - d\varepsilon_s. \quad (1)$$

Typically, the shrinkage strain can be approximated by the following function of time (t):

$$\varepsilon_s = \varepsilon_{s0}(1 - e^{-(t/t_0)}), \quad (2)$$

where ε_{s0} is the final, maximum value of unconstrained shrinkage strain and t_0 is the characteristic time of the polymerization process, i.e. the time taken for the shrinkage strain to reach $(1 - e^{-1}) = 63\%$ of its maximum value. Since changes in the other mechanical properties, i.e. Young's modulus (E) and the viscosity (η), are also controlled by the same polymerization process, for first approximation, we will assume that their changes with time also follow a form similar to that for the shrinkage strain, i.e.

$$E = E_0(1 - e^{-(t/t_0)}) \quad (3)$$

and

$$\eta = \eta_0(1 - e^{-(t/t_0)}). \quad (4)$$

It follows that

$$d\varepsilon_s = \frac{\varepsilon_{s0}}{t_0} e^{-(t/t_0)} dt, \quad (5)$$

$$d\varepsilon_s = \frac{\varepsilon_{s0}}{t_0} e^{-(t/t_0)} dt, \quad (6)$$

and, using a simple Newtonian dashpot,

$$d\varepsilon_e = \frac{d\sigma}{E_0(1 - e^{-(t/t_0)})} \quad (7)$$

where σ is the stress, which is related to the spring stiffness (k), the dimensions (A , l) of the composite sample and the net strain via:

$$d\sigma = -\frac{kl}{A} d\varepsilon. \quad (8)$$

The minus sign is required because a negative strain in the composite produces a tensile force in the spring.

Substituting (5)–(8) into (1), and rearranging gives:

$$\left[\frac{A}{kl} (1 - e^{-(t/t_0)}) + \frac{1}{E_0} \right] \frac{d\sigma}{dt} + \frac{\sigma}{\eta_0} = \frac{\varepsilon_{s0}}{t_0} e^{-(t/t_0)} (1 - e^{-(t/t_0)}). \quad (9)$$

At this point, it would be useful to nondimensionalize the variables as follows:

$$[a(1 - e^{-t'}) + 1] \frac{d\sigma'}{dt'} + b\sigma' = e^{-t'} (1 - e^{-t'}), \quad (10)$$

where $a = AE_0/k = k_0/k$, k_0 being the final effective stiffness of the composite. a can therefore be considered as the compliance of the stress-measuring device relative to that of the composite. $t' = t/t_0$ is the dimensionless time; $\sigma' = \sigma/(E_0\varepsilon_{s0})$ is the ratio between the shrinkage stress and the maximum shrinkage stress that would occur if all the shrinkage strain is converted into elastic strain under the peak Young's modulus; and $b = (E_0t_0)/\eta_0$, as will be seen later, is the inverse of the dimensionless characteristic time that controls the creep rate of the shrinkage stress.

3. Results

3.1. Analytical solutions

To illustrate the evolution of the shrinkage stress and its dependence on the mechanical parameters, we will consider two special cases. The simplifications, with justifications, are also necessary in order to obtain analytical solutions for the differential equation (10).

First, consider the case where the spring stiffness k is infinite, i.e. the compliance ratio $a = 0$. This approximates instruments with feedback control that maintains the sample's thickness, thus giving a zero net strain. Eq. (10) then simplifies to

$$\frac{d\sigma'}{dt'} + b\sigma' = e^{-t'}(1 - e^{-t'}), \quad (11)$$

which, for ease of solving, can be re-expressed as

$$e^{-bt'} \frac{d}{dt'}(e^{bt'} \sigma') = e^{-t'}(1 - e^{-t'}), \quad \frac{d}{dt'}(e^{bt'} \sigma') = e^{-(1-b)t'}(1 - e^{-t'}). \quad (12)$$

Integrating with respect to t' and using the initial condition of $\sigma' = 0$ when $t' = 0$ gives

$$\sigma' = e^{-bt'} \left[\frac{1 - e^{-(1-b)t'}}{1-b} - \frac{1 - e^{-(2-b)t'}}{2-b} \right]. \quad (13)$$

Fig. 2a shows the development of the shrinkage stress with time for the case of zero compliance with different values of b according to (13). As can be seen from these results, b controls the rate of creep in the shrinkage stress: the larger its value, the faster the creep rate. As such, it also controls the maximum value that the shrinkage stress can reach.

For typical dental composites, $E_0 \approx 10$ GPa, $\eta_0 \approx 60,000$ GPa s and $t_0 < 10$ s [15]. b is therefore of the order of 10^{-3} , and can be assumed to be zero. The solution, Eq. (13), for the dimensionless shrinkage stress then becomes:

$$\sigma' = \frac{1}{2}(1 - 2e^{-t'} + e^{-2t'}) = \frac{1}{2}(1 - e^{-t'})^2. \quad (14)$$

The shape of the shrinkage stress curve for $b = 0$ resembles those found experimentally [2,4,7–9,15], which do not show any signs of shrinkage stress decay, at least during the time of measurement. And when Eq. (14) is compared with Eq. (2), it can be seen that the growth rate of the shrinkage stress is slower than that of the shrinkage strain. This also agrees with experiments [2,7,8].

By definition, the elastic strain is

$$\varepsilon_e = \frac{\sigma}{E} = \frac{\sigma' E_0 \varepsilon_{s0}}{E_0(1 - e^{-t'})} = \frac{1}{2} \varepsilon_{s0} (1 - e^{-t'}). \quad (15)$$

As $t' \rightarrow \infty$, $\sigma'_{\infty} \rightarrow 1/2$ or $\sigma_{\infty} \rightarrow (1/2)E_0 \varepsilon_{s0}$ and $\varepsilon_{e\infty} \rightarrow (1/2)\varepsilon_{s0}$. The maximum elastic strain that a dental composite can attain under the zero-compliance condition is therefore half that of the total shrinkage strain. Similarly, the maximum shrinkage stress that it can attain under the same condition is half that it can achieve if all the shrinkage strain is converted into elastic strain with the peak Young's modulus. For typical dental composites, the maximum linear (one-dimensional) shrinkage strain is 0.1–2%, which would give a maximum shrinkage stress of around 5 – 100 MPa for $E_0 = 10$ GPa. Measured shrinkage stress values reported in the literature lie within this range, albeit much closer to the lower end [1–5,7,8].

For example, shrinkage stresses of 5–25 MPa are reported in [1,2,5], and 4–7 MPa in [3,4,8].

In reality, the surrounding constraint has a finite compliance and shrinkage stress is expected to be lower than the upper limit given above. Therefore, we next consider the case where the compliance ratio a is finite but $b = 0$, which is appropriate for dental materials, as shown above. Eq. (10) becomes:

$$\begin{aligned} [a(1-e^{-t'})+1]\frac{d\sigma'}{dt'} &= e^{-t'}(1-e^{-t'}), \\ \frac{d\sigma'}{dt'} &= \frac{e^{-t'}}{(a+1)-ae^{-t'}} - \frac{e^{-2t'}}{(a+1)-ae^{-t'}}. \end{aligned} \quad (16)$$

Using solutions provided by a handbook of integrals [16], together with the same initial condition as in the previous case, it can be shown that

$$\sigma' = \frac{1}{a} - \frac{e^{-t'}}{a} - \frac{1}{a^2} \ln[(a+1)-ae^{-t'}] \quad (17)$$

Fig. 2b shows the build-up of the shrinkage stress with time for different compliance values according to Eq. (17). Note that, with $b = 0$, there is no mechanism to creep out the shrinkage stress. Note also that, in accord with experiments [1,6,7], the larger the compliance ratio, i.e. the more compliant the instrument, the lower is the final shrinkage stress value, which can be found by letting $t' \rightarrow \infty$. Eq. (17) then reduces to

$$\sigma'_\infty = \frac{1}{a} - \frac{1}{a^2} \ln(a+1) \quad \text{and} \quad \sigma_\infty = E_0 \varepsilon_{s0} \left[\frac{1}{a} - \frac{1}{a^2} \ln(a+1) \right] \quad (18)$$

Fig. 3 plots the maximum dimensionless shrinkage stress as a function of the compliance ratio a according to (18). The rapid reduction in the shrinkage stress with increasing compliance of the surrounding constraint can clearly be seen.

Now, $\ln(a+1) \approx a - \frac{a^2}{2} + \frac{a^3}{3} - \dots$. Thus, when $a \ll 1$, $\sigma'_\infty \rightarrow 1/2$ or $\sigma_\infty \rightarrow (1/2)E_0 \varepsilon_{s0}$, which is the same limiting solution obtained previously for the zero-compliance case. This shows internal consistency in the analysis presented, despite the essential simplicity of the model.

3.2. Predictions for stress measurements

Having determined the influence of the external constraint on the shrinkage stress, we proceed to estimate the compliance ratios of some shrinkage stress-measuring systems reported in the literature. This will allow us to calculate the expected shrinkage stresses using Eq. (18) and compare them with experimental measurements.

Some of the shrinkage stress-measuring instruments reported in the literature have a compliance ranging from around 0.1–4 $\mu\text{m/N}$ [4,6,7]. The diameter and thickness of the composite disk samples used in these studies are 3–10 mm and 0.5–1.2 mm, respectively. Using a diameter of 10 mm, thickness 1 mm, and Young's modulus 10 GPa, the compliance of the composite disk is $l/AE_0 = 0.0013 \mu\text{m/N}$. The compliance ratio (a) is therefore around 80–3000, and the corresponding final dimensionless shrinkage stress from Eq. (18) is 3.3×10^{-4} to 0.01. Again, assuming a linear shrinkage strain of 1%, the expected final shrinkage stress is <1 MPa. Fig. 4 shows the predicted shrinkage stress for different disk thicknesses using the above values for material properties and instrument compliance. It can be seen

that, the thicker the disk, the higher the stress. These values and their trends agree very well with those reported in the literature [4,6]. Note that in [4], a correction factor of 4 has been applied to the measured shrinkage stress. When this is removed, the reduced values would agree very well with our predictions. It should also be pointed out that the predicted shrinkage stress is not very sensitive to the Young's modulus value, except at very low instrument compliances.

3.3. Effect of filler fraction

The effect on the shrinkage stress of the filler fraction in the composite has caused much debate, with some experiments producing results that are contrary to expectations [3,5]. It is suspected that differences in the compliance of the equipment may also have contributed to the disagreement.

Eq. (18) for the maximum shrinkage stress for zero creep will be used to consider the combined effect of the filler fraction and the instrument's compliance on the measured shrinkage stress. The following rule of mixtures is used to calculate the composite's elastic modulus [17]:

$$E_0 = \frac{E_r(1+2\chi f)}{1-\chi f}, \quad \chi = \frac{E_f - E_r}{E_f + E_r} \quad (19)$$

where E_r (1 GPa) and E_f (80 GPa) are Young's modulus of the resin and filler, respectively, and f is the filler fraction. The overall composite shrinkage is simply that of the resin multiplied by $(1 - f)$ as the fillers do not shrink. The disk is assumed to be 5 mm in diameter and 2.5 mm thick [3], while the linear shrinkage strain of the resin is assumed to be 2.6% [17].

Fig. 5 shows the relationship between the shrinkage stress and the filler fraction for different instrument compliances. The range of compliance values considered covers those of shrinkage stress-measuring equipment as well as composite-restored teeth; see later. Again, it can be seen that the higher the compliance of the surrounding constraint, the lower the level of shrinkage stress. It can also be seen that the shrinkage stress initially increases with increasing filler fraction, a trend similar to those reported in [3,5]. After reaching a peak value, it then reduces quickly with further increases in the filler content. The more compliant the surrounding constraint, the earlier the increased filler content will be capable of reducing the shrinkage stress. The latter agrees with the observation that when instrument with a high compliance is used, materials with lower shrinkage strain, because of a high filler fraction, produces lower shrinkage stress despite higher Young's modulus [4].

3.4. Compliance of a model tooth cavity

While the mathematical model developed above is only one-dimensional, the concept of the compliance ratio should be universal. Thus, using a geometrically simple model, we consider the compliance ratio of a Class-I cavity, which is considered to have the most confined configuration of all cavities. The said cavity is simplified as a thick-walled cylinder (Fig. 6), with the 3-D effect presented by the floor being ignored.

Consider first the stiffness per unit area, k_c , of the composite restoration in Fig. 6. By definition,

$$k_c = \frac{\sigma_r}{u} \bigg|_{r=r_i}, \quad (20)$$

where σ_r is the radial stress, u is the radial displacement and r_i is the inner radius of the cavity or, simply, radius of the restoration. Under a uniform tensile stress, σ , $\sigma_r = \sigma_\theta = \sigma$ everywhere within the composite, where σ_θ is the circumferential stress. Using Hooke's law, the circumferential strain is

$$\varepsilon_\theta = \frac{1}{E_c}(\sigma_\theta - \mu_c \sigma_r) = \frac{1 - \mu_c}{E_c} \sigma. \quad (21)$$

where E_c and μ_c are Young's modulus and Poisson's ratio of the composite, respectively. The circumferential strain is related to the radial displacement u by $\varepsilon_\theta = u/r$. Substituting (21) into (20) gives:

$$k_c = \frac{E_c}{r_i(1 - \mu_c)}. \quad (22)$$

Consider next the stiffness per unit area, k_t , of the surrounding tooth tissue in Fig. 6. The same stress σ acts on the inner wall of the cavity. Using Lamé's equations for thick-walled cylinders and the boundary conditions of $\sigma_r = \sigma$ at $r = r_i$ and $\sigma_r = 0$ at $r = r_0$, the outer radius, it can be shown that the circumferential strain at the inner wall is

$$\varepsilon_\theta = -\frac{\sigma}{E_t} \left[\frac{1 - \mu_t}{R^2 - 1} + \frac{R^2(1 + \mu_t)}{R^2 - 1} \right]. \quad (23)$$

where E_t and μ_t are Young's modulus and Poisson's ratio of the tooth, respectively, and R is the ratio between the outer and inner radii.

Using formulae similar to those used for the composite and noting that a tensile stress gives rise to negative displacement, the stiffness per unit area, k_t , of the tooth cavity can be shown to be:

$$k_t = \frac{E_t}{r_i \left[((1 - \mu_t)/(R^2 - 1)) + ((R^2(1 + \mu_t))/(R^2 - 1)) \right]}. \quad (24)$$

It follows that the compliance ratio of the model composite-restored tooth is

$$a = \frac{k_c}{k_t} = \frac{E_c}{E_t} \frac{[(1 - \mu_t) + R^2(1 + \mu_t)]}{(1 - \mu_c)(R^2 - 1)}. \quad (25)$$

Assuming $R = 2$, $\mu_t = \mu_c = 0.2$, $E_c = 10$ GPa and $E_t = 80$ GPa (enamel's modulus), the compliance ratio of the composite-restored tooth has a value of 0.29. From Fig. 3, this will give a maximum dimensionless shrinkage stress of around 0.4, i.e. an actual final shrinkage stress of 40 MPa, based on the same material constants used previously. Given that the bond strength between composite restorations and tooth tissues is around 20 MPa, it is perhaps not surprising that interfacial debonding has been observed in some composite restorations [18]. It should be pointed out that a much lower shrinkage stress will be obtained if properties of the softer dentin are used instead, or if a Class-II MOD restoration is considered [6].

4. Discussion

A mathematical model has been presented in this paper to describe the shrinkage stress kinetics in dental composites. Application of the analytical solutions to shrinkage stress-measuring instruments with different compliances has provided very good agreement between theory and experiment in terms of the temporal development and final value of the shrinkage stress. The model correctly and precisely predicts the increase in shrinkage stress with reduced instrument compliance and increased disk thickness [1,4,6,7]. It also correctly predicts the effect of the filler fraction on the shrinkage stress measured by instruments with different compliances [3–5].

The model provides an explicit expression, Eq. (18), linking the maximum shrinkage stress with the compliance and material properties of the entire system. Note that the compliance ratio a is a function of the elastic modulus of the composite as well as the instrument compliance. Therefore, Eq. (18) captures fully the interaction between these two factors in determining the maximum shrinkage stress value. With this expression, the effect of the filler fraction of the composite and its interaction with the compliance of the surrounding constraint can be studied explicitly and comprehensively (Fig. 5).

It is considered by many that increasing the filler content will reduce the shrinkage stress. The current theory shows clearly that this is not necessarily the case. The initial increase in shrinkage stress with increasing filler fraction (Fig. 5) has been seen in experimental results reported by others [3,5]. This is attributed to the rapid increase in Young's modulus with filler content. Fig. 5 shows that the potential to reduce shrinkage stress by increasing the filler content depends to a great extent on the compliance of the external constraints, with the method being more effective with high-compliance restoration. Therefore, when choosing an appropriate filler fraction with the aim of ensuring that the shrinkage stress does not exceed its critical value, one would first need to estimate the compliance of the surrounding constraint. The current theory also shows that the eventual reduction in shrinkage stress with further increase in filler content is due to a rapid increase in the compliance ratio, as a result of the stiffening of the composite, as well as a reduction in shrinkage strain.

The two dimensionless parameters, a and b , are the main features of the current model. a , the compliance ratio, describes the relative compliance between the external constraint and the composite sample itself. Its effect on the shrinkage stress is similar to that of the C -factor (the ratio between the bonded to unbonded surfaces). However, the compliance ratio takes into account both the geometrical (diameter, thickness) and material (Young's modulus, Poisson's ratio) factors. It is therefore a fuller description of the mechanical behavior of the system. More importantly, the theory suggests that, in considering shrinkage stress development, the compliance of the whole system needs to be considered. In the case of shrinkage stress measurement using an instrument, one would need to include the connectors linking the composite sample to the instrument, as well as the instrument itself. Similar results showing the importance of instrument compliance on shrinkage stress measurement have been demonstrated by others using finite element analysis [12,13].

We are now in a position to understand the different correlations between the shrinkage stress and the material properties and disk dimensions reported in the literature. For instruments with a very low compliance, high correlations have been reported between the shrinkage stress, shrinkage strain and Young's modulus [5,7], i.e. there are strong linear relationships between these variables. On the other hand, when the instrument compliance is high, such a linear relationship was not observed between the shrinkage stress and Young's modulus [7]. This apparent inconsistency in the shrinkage stress response of composites can

again be explained using the current model. Recall that when the compliance ratio a is small, the shrinkage stress $\sigma_{\infty} \rightarrow (1/2) E_0 \varepsilon_{s0}$, which clearly indicates strong linear relationships that are independent of the sample dimensions between the shrinkage stress, the shrinkage strain and Young's modulus. This agrees with the asymptotic behavior of shrinkage stress with increasing specimen thickness, or compliance, reported in [19]. On the other hand, when a is large, $(1/a) \gg (1/a^2) \ln(a+1)$. Eq. (18) then reduces to $\sigma_{\infty} = (E_0 \varepsilon_{s0})/a = (\varepsilon_{s0} k l)/A$, which agrees with the observation reported in [7] that "when more compliance was allowed, measured stress was highly related to shrinkage strain, and not elastic modulus." It also agrees with the increase in shrinkage stress with increased disk thickness reported in [6], the reduction in shrinkage stress with increased disk diameter reported in [19], and the linear relationship between spring stiffness and shrinkage stress reported in [1]. Note that the compliance of the remaining cusps of a Class-II cavity has been measured to be around 3 $\mu\text{m/N}$ [6], which supports the use of compliant measurement systems.

While its effect seems to be secondary, at least in the short term because of its small value for dental composites, the decay constant $b = (E_0 t_0)/C_0$ encapsulates both the mechanical (Young's modulus, viscosity) and chemical (characteristic time of the polymerization shrinkage) characteristics of the composite. t_0/b is the characteristic time for shrinkage stress decay. Using typical values for dental composites, it is estimated to be in the region of hours. For systems that have a faster rate of decay in the shrinkage stress, b may play a bigger role in the characterization and design of the material as it also controls the maximum value of the shrinkage stress that can be achieved (see Fig. 2a). Besides, for the long-term integrity of a composite restoration, for which fatigue failure would be more relevant, it is important to minimize any pre-stress in the system. It is therefore of benefit to creep out the shrinkage stress as early as possible.

While reasonable for shrinkage strain, it may not be accurate to use the same exponential form for the growth in Young's modulus and viscosity with time during polymerization. Experiments have shown a slower initial rate of growth for these mechanical properties [7,10]. This is because the cross-linking of the polymeric chains which gives the material its stiffness and viscosity occurs later in the polymerization process. This may explain why the predicted stresses are higher than those reported in the experimental studies. In general, it can be assumed that

$$E = E_0 (1 - e^{-(t/t_0)})^n \quad (26)$$

where $n > 1$. The greater the value of n , the slower is the rate of increase in the mechanical properties with time. Further, it can be shown that, for the case of zero creep and zero compliance, the maximum dimensionless shrinkage stress is $\sigma'_{\infty} \rightarrow 1/(n+1)$. Hence, a more effective way to reduce shrinkage stress is perhaps to delay the formation of cross-linking between the polymeric chains. Indeed, in addition to its low shrinkage strain, the low shrinkage stress found in the silorane composite can also be attributed to its slow rate of modulus development [7].

The current model suggests that there is a limit to the maximum shrinkage stress that can be generated, even in the absence of creep and system compliance, and it is only a fraction of the product of the final Young's modulus and shrinkage strain. As shown above, the exact value of the maximum shrinkage stress depends on the kinetics of the shrinkage strain and modulus development. This may seem surprising. After all, if there is no creep, should not all the shrinkage strain be converted into elastic strain and the final shrinkage stress be equal to $E_0 \varepsilon_{s0}$? To answer this, one should remember that during the early stage of polymerization, the composite is rather soft. It is therefore not capable of stretching the

instrument to produce the maximum stress possible in itself. By the time it becomes fully rigid (post-gel), the strain locked in would be smaller than the free shrinkage strain. This is why the final shrinkage stress is lower than $E_0\varepsilon_{s0}$. It follows that the composite will undergo permanent deformation under constrained shrinkage even in the absence of creep. This can be seen from the final elastic strain, which is predicted to be only a fraction of the total shrinkage strain. Therefore, upon unloading, the total shrinkage strain will not be fully recovered, and the sample will be longer than the one that has undergone free shrinkage. This behavior is similar to that of a plastic material undergoing strain-hardening, with the polymeric cross-linking performing a similar role to that of the dislocations in metals. In [5], a flow factor for resin composites is defined as $(\sigma_{th} - \sigma_{\infty})/\sigma_{th}$, where $\sigma_{th} = E_0\varepsilon_{s0}$. The average value found using a zero-compliance instrument for 17 different composites is around 60%. Using our current result of $\sigma_{\infty} = (1/2) E_0\varepsilon_{s0}$, the flow factor will be 50%. Note that even better agreement will be achieved if $n > 1$ is used in the calculation (see Eq. (26)).

The analytical solutions presented herein are only applicable to simple geometries with uniform stress distributions. In reality, composite restorations have complex geometries and highly nonuniform stress distributions. To analyze them, a numerical approach such as the finite element method would be required. Thermal strains are ignored in this study. To consider them, we would need to know the temporal changes in temperature during curing and the corresponding changes in the coefficient of thermal expansion. However, experiments seem to indicate that their effect on the shrinkage stress/strain is not significant.

5. Concluding remarks

The current study represents the most comprehensive theoretical treatment to date of the subject of polymerization shrinkage stress in dental composites. The mathematical model correctly predicts how shrinkage stress increases with time, its dependence on the interaction between the entire system's compliance and the material properties, and the effect of the filler fraction on its maximum value. The analytical solutions allow us to understand the myriads of experimental results reported in the literature and explain their apparent differences. They can also provide some useful guidelines for optimizing the mechanical performance of dental composite restorations, which needs to be considered in conjunction with the compliance of the whole restoration.

Acknowledgments

The author would like to thank Drs. Jianying Li and Haiyan Li for some useful discussion. This study was supported in part by the National Institute of Health through Grant 1R01DE021366-01.

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Fig. 1. Model for shrinkage stress-measuring system showing connection between instrument and composite sample with thickness l and diameter D .

Fig. 2.
 (a) Shrinkage stress vs. time for the case of zero compliance ($a = 0$) with different b values.
 (b) Shrinkage stress vs. time in the absence of creep ($b = 0$) with different compliance values.

Fig. 3.
Dependence of maximum shrinkage stress on compliance ratio in the absence of creep ($b = 0$).

Fig. 4.
Predicted shrinkage stress as a function of instrument compliance and specimen thickness.

Fig. 5.
Effect of filler fraction on shrinkage stress measured by instruments with different compliances.

Fig. 6. Thick-walled cylinder model for a tooth with a Class-I composite restoration. Both restoration and tooth are subjected to a tensile stress σ .