Quantitative determination of stress reduction by flow in composite restorations

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Abstract—In this study, the reduction of the polymerization shrinkage stress by flow of four chemically-initiated composites was investigated in relation to the cavity configuration. In an experimental set-up simulating restorations bonded to cavity walls, the developing shrinkage stress accompanied by flow was recorded as a function of time for several configurations. For each configuration, theoretical shrinkage stress curves were also drawn, excluding stress reduction by flow. These data were obtained from Young's modulus determinations at the early setting stage and the corresponding polymerization shrinkage. By comparison of the theoretical stress with the experimentally determined stress, a measure for the ability to flow in the bonded situation could be obtained. It was found that the flow strongly depended on the type of composite and on the configuration of the cavity.

The polymerization contraction of composites used in adhesive restorations induces stress, which is associated with stiffness of the restorative material as well as with that of the restored tooth. This stress diminishes by (plastic) flow (Davidson and De Gee, 1984), which partly accommodates to the polymerization shrinkage.

During curing of a bonded composite restoration, a complicated process takes place: Its restrained shrinkage induces stress which, in its turn, if the stress exceeds the elastic limit, induces plastic deformation. During the early stage of setting, the resin network is still weak and therefore the elastic limit will be low. Plastic yielding to the stress at this stage of setting can be achieved without damage of the internal structure of the resin composite and the adhesive bond, since the molecules can still slip into new positions and orientations. This kind of deformation can be characterized as flow. When the curing proceeds, contraction and flow decrease gradually, while stiffness increases. As a result, the stress will still grow with time and may cause serious problems for the maintenance of the adhesive bond or may even cause cohesive failure of the restorative material or the surrounding tooth tissue (Eick and Welch, 1986; McCullock and Smith, 1986; Kemp-Scholte and Davidson, 1988).

Since the ability of a bonded composite to deform elastic and/or plastically is configuration-dependent, the magnitude of polymerization contraction stress will also be configuration-dependent (Feilzer *et al.*, 1987). Therefore, it is not possible to determine the flow as function of time as an intrinsic material property. Only for given configurations will the material produce corresponding stress values and thus also related flow-controlled stress relaxation.

The purpose of this study was to establish the actual stress reduction by flow of composite restorations at various cavity configurations by comparison of a calculated shrinkage stress, in which release by flow is excluded, with experimental stress, in which some reduction by flow always occurs (Davidson and De Gee, 1984).

MATERIALS AND METHODS

The materials used in this study are listed in Table 1.

To assess the amount of flow of chemically-initiated composites bonded to the walls of cavities with various configurations, we used an experimental set-up which was similar to that used in a previous study (Feilzer *et al.*, 1987). In short, the experimental set-up consisted of two opposing identical steel disks (ϕ = 10.0 mm), between which freshly mixed composite was inserted and shaped to a cylinder according to the circumference of the disks. Schematically, this represented a filled cavity of which the configuration could be chosen by varying the diameter (d) of the disks and/or their mutual distance (h). Each configuration was fixed by its so-called "C-factor", being the ratio between the restoration's bonded surface (disk areas) and the free unbonded surface (cylinder jacket) (c = d/2 h). In this investigation, configurations with C-factors of 0.5, 2.0, 2.5, and 5.0 were studied. These were obtained with disks with a diameter of 10.0 mm and disk-to-disk distances of 10.0 mm, 2.5 mm, 2.0 mm, and 1.0 mm, respectively. Bonding was established by silane-coating of the disk surfaces (Kulzer & Co. GmbH, Silicoater, FRG). One disk was connected to the load-cell and the other to the crosshead of a tensometer (Instron 6022, Instron Limited, England) which continuously counteracted the yielding of the load-cell to the shrinkage

TABLE 1
THE COMPOSITES USED IN THIS STUDY

Brand	Batch	Manufacturer
P10	A: 7AE1 B: 7AE1	3M, St. Paul, MN
Silar	A: 7C3 B: 7E3	3M
Brilliant	u/c 120882-04	Coltène, Altstätten, Switzerland
Clearfil Posterior	KC0107 KU0207X	Cavex, The Netherlands

TABLES 2-4
POLYMERIZATION SHRINKAGE, YOUNG'S MODULUS, EXPERIMENTAL AND THEORETICAL POLYMERIZATION CONTRACTION STRESS AT C = 0.5 FOR SILAR, BRILLIANT, AND CLEARFIL POSTERIOR (SD)

(2) Silar

time (min)	Pol. shrinkage (vol%)	Young's modulus (MPa)	σ_{th} (MPa)	σ_{exp} (MPa)
3′	0.5 (0.2)	7 (7)	0	0
4′	1.4 (0.5)	391 (231)	1.4	0.2 (0.2)
5′	1.6 (0.3)	1081 (482)	2.3	0.6 (0.2)
6′	1.8 (0.2)	1619 (434)	3.6	1.3 (0.4)
7′	2.0 (0.2)	1977 (362)	5.2	1.8 (0.5)
8′	2.1 (0.2)	2543 (437)	6.2	2.3 (0.5)
10′	2.2 (0.3)	2870 (503)	7.3	2.7 (0.6)
15′	2.3 (0.2)	3194 (406)	8.6	3.3 (0.6)
20′	2.4 (0.3)	3701 (816)	10.1	3.6 (0.6)
30′	2.4 (0.2)	4099 (950)	10.1	3.9 (0.6)
60′	2.5 (0.2)	4359 (721)	11.8	4.1 (0.7)

(3) Brilliant

time (min)	Pol. shrinkage (vol%)	Young's modulus (MPa)	σ_{th} (MPa)	σ_{exp} (MPa)
3′	0.6 (0.3)	606 (401)	1.5	0.2 (0.1)
4′	1.4 (0.3)	1726 (499)	7.0	1.3 (0.3)
5′	2.0 (0.2)	2310 (755)	12.5	2.6 (0.3)
6′	2.5 (0.2)	3810 (252)	20.1	3.3 (0.2)
7′	2.8 (0.2)	4019 (761)	25.0	3.9 (0.1)
8′	3.0 (0.2)	4879 (593)	28.9	4.3 (0.1)
9′	3.2 (0.2)	5672 (65)	33.4	4.5 (0.1)
10′	3.3 (0.2)	6186 (379)	35.9	4.7 (0.0)
15′	3.6 (0.2)	7557 (554)	44.9	5.3 (0.0)
20′	3.7 (0.2)	8364 (68)	48.3	5.6 (0.1)
30′	3.9 (0.2)	9591 (914)	56.0	5.9 (0.2)
60′	4.2 (0.3)	11022 (353)	69.2	6.4 (0.1)

(4) Clearfil Posterior

time (min)	Pol. shrinkage (vol%)	Young's modulus (MPa)	σ_{th} (MPa)	σ_{exp} (MPa)
3′	-	3 (2)	0.0	0.0
4′	0.4 (0.2)	14 (9)	0.0	0.0
5′	0.9 (0.2)	121 (165)	0.3	0.0
6′	1.4 (0.2)	341 (404)	0.9	0.3 (0.2)
7′	1.9 (0.2)	788 (619)	2.5	0.6 (0.5)
8′	2.3 (0.2)	1785 (1083)	5.4	1.1 (0.5)
9′	2.4 (0.2)	2617 (583)	6.4	1.4 (0.5)
10′	2.7 (0.2)	3743 (1145)	10.9	1.9 (0.8)
15′	3.4 (0.1)	7310 (863)	31.4	3.1 (0.5)
20′	3.6 (0.1)	9315 (1513)	38.8	3.8 (0.4)
30′	3.9 (0.2)	11819 (2363)	53.0	4.5 (0.3)
60′	4.2 (0.5)	13188 (1126)	68.8	5.3 (0.3)

force to maintain the original disk-to-disk distance very accurately. Under these restricted conditions (simulating restorations in a bonded situation), the development of the polymerization shrinkage stress, σ_{exp}, which is attendant with flow, was recorded continuously (at RT = 23°C) and can be given by: $\sigma_{exp} = \sigma_{th} - \sigma_{red}$. σ_{th} denotes the calculated theoretical stress, which would develop if stress reduction by flow (σ_{red}) were excluded. σ_{red}, therefore, indirectly represents a measure for flow and can be calculated from $\sigma_{th} - \sigma_{exp}$. The theoretical stress, σ_{th}, can be expressed by:

$$\sigma_{th} = \int_0^t E(t)d\epsilon(t) \qquad (1)$$

where E is Young's modulus at time t, and $d\epsilon$ an infinitely small elastic elongation of the sample to its original length, equal to the increase of axial shrinkage (within a period dt), which would occur at time t, *under free shrinking conditions*. According to Feilzer *et al.* (1989), the free axial polymerization shrinkage of a resin, bonded between two opposing surfaces, depends on its C-factor. Therefore, corrections have to be made. For C = 0.5, 2.0, 2.5, and 5.0, studied in this investigation, the free axial shrinkage is approximately 0.40, 0.60, 0.65, and 0.85 times the (free) volumetric shrinkage (%).

The experimental procedure to determine E was carried out simultaneously with the determination of σ_{exp}, and consisted of periodical cycling of the cross-head up and down (speed 0.05 mm/min) around the instantaneous disk distance with a strain of 0.01%. From each cycle taken at time t, Young's modulus is given by: $(\sigma_{u,t} - \sigma_{1,t}) \cdot 10^4$, in which $\sigma_{u,t}$ and $\sigma_{1,t}$ are the highest and lowest stress, corresponding with upper and lower positions (10^{-4} rel. strain) of the cross-head at time t. Although this experiment allowed Young's modulus to be calculated every 2.4 s, for practical reasons, moduli were calculated at only a limited number of time periods, *i.e.*, from cycles taken at t = 3, 4, 5, 6, 7, 8, 9, 10, 15, 20, 30, and 60 min from the start of mixing. The theoretical stress can then be expressed by the approximation:

$$\sigma_{th} = \sum_{t=0}^{t} E_t \cdot \epsilon_t \qquad (2)$$

where E_t represents Young's modulus at one of these time periods and ϵ_t the increase of free axial polymer-

TABLE 5
POLYMERIZATION SHRINKAGE, YOUNG'S MODULUS, EXPERIMENTAL AND THEORETICAL POLYMERIZATION CONTRACTION STRESS AT C = 0.5, 2.0, 2.5, AND 5.0 FOR P10 (SD)

time (min)	Pol. shrinkage (vol%)	Young's modulus (MPa)	σ_{th} C = 0.5	σ_{exp} (MPa)	σ_{th} C = 2.0	σ_{exp} (MPa)	σ_{th} C = 2.5	σ_{exp} (MPa)	σ_{th} C = 5.0	σ_{exp} (MPa)
3'	-	2 (3)	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
4'	0.4 (0.2)	19 (9)	0.0	0.0	0.0	0.2 (0.0)	0.0	0.1 (0.1)	0.1	0.2 (0.1)
5'	0.9 (0.3)	175 (106)	0.4	0.2 (0.1)	0.6	1.3 (0.2)	0.6	1.1 (0.0)	0.8	1.1 (0.2)
6'	1.2 (0.3)	893 (347)	1.5	0.4 (0.2)	2.2	3.0 (0.5)	2.4	2.8 (0.4)	3.1	3.4 (0.8)
7'	1.6 (0.2)	1313 (573)	3.6	1.0 (0.3)	5.3	4.4 (0.4)	5.8	5.2 (0.4)	7.5	6.6 (1.0)
8'	1.8 (0.1)	2760 (405)	5.8	1.6 (0.6)	8.6	6.8 (0.7)	9.4	7.0 (1.6)	12.2	8.1/
9'	1.9 (0.1)	3614 (977)	7.2	2.2 (0.7)	10.8	8.1 (1.0)	11.7	8.3 (2.1)	15.3	fracture
10'	2.0 (0.1)	4798 (1397)	9.1	2.6 (0.8)	13.6	8.9 (1.0)	14.8	11.3/	19.7	
15'	2.2 (0.1)	8429 (1612)	15.8	4.8 (1.3)	23.8	11.8 (1.1)	25.7	fracture	33.7	
20'	2.4 (0.1)	11085 (932)	24.7	5.6 (1.6)	37.1	13.2 (0.7)	40.2		52.6	
30'	2.6 (0.1)	16522 (2190)	38.0	6.3 (1.3)	56.9	14.4 (0.5)	61.6		80.7	
60'	2.8 (0.2)	20290 (499)	54.2	7.4 (1.9)	81.3	14.8 (0.2)	88.0		115.1	

ization shrinkage during the period from one cycle to the next, taken at time t.

The (free) volumetric polymerization shrinkage was determined during a period of 60 min at 23 ± 0.1°C with the modified mercury dilatometer, as described by Feilzer *et al.* (1988). Both the development of the experimental stress σ_{exp} and Young's modulus E were determined for the configuration factor C = 0.5, also during a period of 60 min. From these data, the theoretical stress σ_{th} and the stress reduction by flow σ_{red} were calculated. In addition, σ_{exp}, σ_{th}, and σ_{red} were also determined for P10 at C = 2.0, 2.5, and 5.0, in which σ_{th} was calculated from E values found for C = 0.5. All experiments were repeated at least three times and averaged.

RESULTS

Tables 2-5 compile the data at the time periods 3, 4, 5, 6, 7, 8, 9, 10, 15, 20, 30, and 60 min of the experimental polymerization shrinkage stress σ_{exp}, volumetric polymerization shrinkage, and Young's modulus, together with the theoretical stress σ_{th}. The graphical representation of σ_{exp} and σ_{th} is shown in Figs. 1-3.

DISCUSSION

In the experimental set-up in which the axial polymerization shrinkage of the cylindrical samples was continuously counteracted to maintain the original length, the samples were in fact continuously elongated to compensate for the polymerization shrinkage. The tensile stress which developed accordingly increased at each time increment by an amount which was determined from the strain needed for the shrinkage compensation during that time increment and the respective Young's modulus. Since this process was always accompanied by flow to relieve the developing tensile stress, the axial shrinkage to be compensated for was at any time smaller than that which would have taken place under free shrinking conditions (Feilzer *et al.*, 1989). The theoretical approach takes this into account, *i.e.*, the stress increments were calculated from shrinkage compensations based on the free axial shrinkage. Although the theoretical stress curves may be calculated more accurately from a large number of small increments, a reasonable approximation was ob-

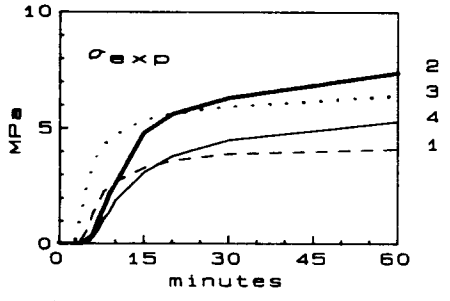

Fig. 1. Measured polymerization contraction stress, at C = 0.5, for Silar (1), P10 (2), Brilliant (3), and Clearfil Posterior (4).

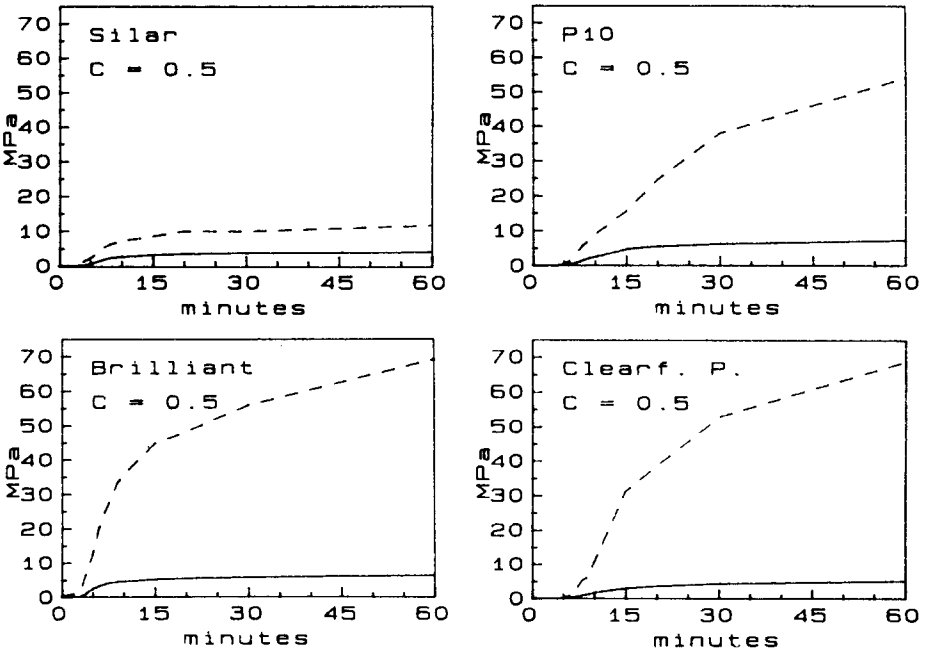

Fig. 2 a,b,c,d. Measured polymerization contraction stress, together with the theoretical polymerization contraction stress, in which flow is excluded, for P10, Silar, Brilliant, and Clearfil Posterior, at C = 0.5.

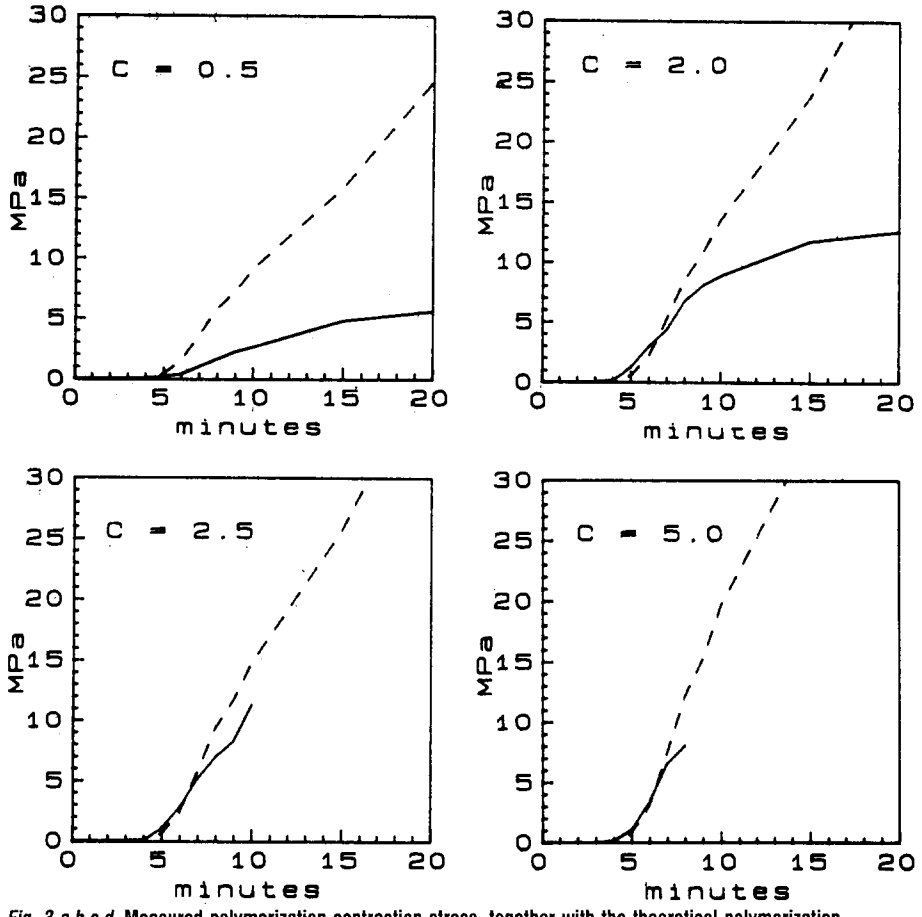

Fig. 3 a,b,c,d. **Measured polymerization contraction stress, together with the theoretical polymerization contraction stress, in which flow is excluded, for P10, at C = 0.5; C = 2.0; C = 2.5; and C = 5.0, respectively.**

tained with the chosen set of time increments. In a previous study (Davidson and De Gee, 1984), only one large time interval was used for the calculation of the theoretical stress, which explains the significantly higher values of σ_{th} (at 45 min) in that study.

The justification of the method presented for the determination of Young's modulus at the early setting of the resin composite was based on the interpretation of the plots obtained from the cycling around the original sample height. For each cycle of 2.4 sec and total strain of 0.01%, the stress at passing through and returning to the original sample height was always equal to or slightly higher than that at the start. From the linearity of the stress-strain curve, the deformation could be regarded exclusively as elastic strain, from which Young's modulus was calculated.

Fig. 1 shows that the development of the experimental stress σ_{exp}, for different product samples with equal configuration (C = 0.5), differed significantly. Brilliant developed fastest, Clearfil Posterior slowest. Although slow development can be regarded as an advantage, because this allows more time for the bonding agent to mature to full strength (Davidson *et al.*, 1984; Braem *et al.*, 1987), none of the tested composites developed a polymerization contraction stress at C = 0.5, which exceeded the bond strength of presently available dentin bonding agents (Kemp-Scholte and Davidson, 1990).

In Fig. 2 and Fig. 3, the vertical distance between dotted and solid curves $\sigma_{th} - \sigma_{exp}$ can be considered as stress reduction by flow σ_{red}. Cohesive micro-fracturing contributing to the reduction is not likely to occur, since it was shown that the ultimate tensile strength of composite contracting under restricted conditions was not inferior to that of free shrinking materials (Davidson and De Gee, 1984).

For comparison of the stress-reduction-by-flow properties of different composite materials, the relative stress reduction was defined by $f = \sigma_{red}/\sigma_{th} \times 100\%$. In addition, since the stress reduction by flow was shown to depend on the configuration (Fig. 3), f must be specified together with the C-value of application. When the materials were compared at 60 min after initiation, at a configuration factor of C = 0.5, the relative stress reductions ($f_{c=0.5}$) for Clearfil Posterior, Brilliant, P10, and Silar were 92%, 91%, 86%, and 65%, respectively. Silar has a low calculated polymerization contraction stress, which must be attributed to its low Young's modulus. Therefore, the required stress reduction will be limited, and f also appeared to be low.

At a low C-value, the stress can be built up without interference by fracture, but for higher C-values, failure of the materials might result from insufficient flow capacity (Feilzer *et al.*, 1987). The decreasing of f with increasing C-factor is demonstrated for P10 in Fig. 3. For samples with a configuration factor of C = 0.5, at 10 minutes' setting, 71% of the polymerization stress was relieved by flow, while at C = 2.0 this was only 35%. For samples with a configuration factor C = 2.5 or C = 5.0, the stress reduction by flow of the sample (respectively, 24% and negligible) was not high enough to prevent fracture, since, ultimately, flow capacity and strain capacity were exceeded. It can be concluded that: (1) stress reduction by flow and flow capacity is material- as well as configuration-dependent, and (2) flow of a composite restorative material contributes greatly to reduction of the shrinkage stress in restorations with a low C-factor (large free unbonded surface), and insufficiently in situations with high C-factors.

Although no sufficient data are available for light-curing composite, it can be expected that due to the fast development of the polymerization reaction, the stress reduction by flow is much more restrained than for chemically-initiated materials. Moreover, in most clinical situations, the surface exposed to the light source which could provide material for flow is the one which sets first. Therefore, preserving bonding to tooth structure might be more dif-

ficult with light-initiated than with chemically-initiated composite. Appreciation of flow capacity and flow direction is of great importance to the general practitioner in achieving more successful restorations. In spite of its relatively complicated characterization, this property certainly deserves attention in composite specification.

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