

Thermal Expansion of Composite Resins and Sealants*

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Linear thermal coefficients of expansion (α) of seven commercial composite resins and four pit and fissure sealants were determined between 0 and 60°C on a thermomechanical analyzer. The thermal expansion curves obtained were nonlinear. Values of α ranged from 26.5 to 39.6 $\times 10^{-6}$ °/C for the composite and from 70.9 to 93.7 $\times 10^{-6}$ °/C for the sealants.

J Dent Res 58(2):584-587, February 1979

Introduction.

Values of the linear thermal coefficient of expansion (α) of composite restorative materials have been reported by Macchi and Craig¹, and subsequently by Dennison and Craig², in their evaluations of physical and mechanical properties. In these studies, thermal dimensional change was determined on a specimen (6.4 mm square and 51 mm long) between 24 and 88 C with a method similar to that described in ASTM Specification D-696 for plastics.³ Values of α , for composite resins available,^{1,2} ranged from 27 to 41 $\times 10^{-6}$ °/C.

The purpose of this investigation was to determine the linear thermal coefficient of expansion of seven commercial composite resins and four pit and fissure sealants between 0 and 60°C from thermal expansion curves of specimens smaller than previously studied.

Received for publication February 28, 1978

Accepted for publication April 12, 1978

*This investigation was supported by Biomedical Research Support Grant RR-05321 from the National Institutes of Health, Bethesda, MD 20014.

The cooperation of the following companies in providing commercial products is acknowledged: L. D. Caulk Co., Johnson & Johnson, Kerr Mfg. Co., 3M Co., and S. S. White.

This investigation was presented at the annual meeting of the International Association for Dental Research in Washington, D.C., March, 1978.

Materials and methods.

Seven commercial composite resins and four commercial pit and fissure sealants were evaluated for thermal expansion. Codes, batch numbers, and manufacturers of the products studied are listed in Table 1.

Five cylindrical specimens (3.7 mm in diameter and 7.7 mm long) were made for each product by polymerizing the resin in a metal die. The specimens were placed in an oven at 37°C within 90 seconds after initiating the mix, and were stored dry for 24 hours before testing. Specimens of the two materials activated by ultraviolet light (NF and NS) were polymerized in borosilicate glass tubing (3.50 mm inside diameter) by exposing the material to an ultraviolet light source* for a total of 20 minutes at a distance of 5 cm.

Thermal expansion was measured by a commercial thermomechanical analyzer (TMA)† at a heating rate of 5°C/minute. A quartz probe (2.5 mm in diameter), which is attached to a metallic sleeve that serves as the core of a linear variable transformer (LVDT), was mechanically positioned by a spring on the surface of a specimen. The output of the LVDT was then balanced and zeroed electrically so subsequent vertical displacement could be calibrated and recorded as a function of temperature. A schematic sketch of the TMA cell has been shown previously.⁴ The probe was calibrated by measurement of the thermal expansion of an aluminum sample† with a linear coefficient of thermal expansion (α) of 23.2 $\times 10^{-6}$ °/C at a temperature of 27°C. The mean calibration factor (S), determined from five replications, was 2.61 $\mu\text{m}/\text{cm}$ of chart paper with a standard deviation of 0.05 $\mu\text{m}/\text{cm}$. The linear coefficient of thermal expansion (α) was related

*Blak-Ray Model B-100A, Ultra-Violet Products, Inc., San Gabriel, CA 91775.

†duPont 941 Thermomechanical Analyzer, E. I. duPont de Nemours & Co., Inc., Instrument Products Division, Wilmington, DE 19898.

TABLE 1
CODE, BATCH NUMBERS AND MANUFACTURERS OF PRODUCTS TESTED.

CODE	PRODUCT NAME	BATCH NUMBERS	MANUFACTURER
Composite Resins:			
A	Adaptic	base-7E019 catalyst-7E019	Johnson & Johnson Dental Products Division East Windsor, NJ 08520
AR	Adaptic Radiopaque	base-6L103 catalyst-6L103	Johnson & Johnson
C	Concise	base-7080E12 catalyst-7080E12	3M Co. St. Paul, MN 55101
NF	Nuva-Fil	base-052877 activator-120276	L. D. Caulk Co. Division of Dentsply International, Inc. Milford, DE 19963
P	Powderlite	powder-097702 liquid-027612	S. S. White Division Pennwalt Corporation Philadelphia, PA 19102
S	Simulate	base-0771061 catalyst-772096	Kerr Manufacturing Co. Division of Sybron Corp. Romulus, MI 48174
V	Vytol	base-042077 catalyst-050477	L. D. Caulk Co.
Sealants:			
D	Delton	universal-1047D202 catalyst-1047D202	Johnson & Johnson
K	Kerr Sealant	base-0513771083 catalyst-0513771083	Kerr Manufacturing Co.
NS	Nuva-Seal	base-040477 initiator-120276	L. D. Caulk Co.
W	White Sealant System	resin A-70702 resin B-7059B1	3M Co.

to the calibration factor (S) by the equation: $S = \alpha h / (\Delta l / \Delta T)$, where h is the original height of the specimen and $(\Delta l / \Delta T)$ is the slope of the thermal expansion curve measured as the tangent to the curve at a given temperature, or determined as the straight line between two temperatures.

Thermal expansion curves were obtained over a temperature range of 0 to 60°C with the quartz probe applying a pressure of 2.0 kPa on dry specimens. To obtain a uniform heating rate initially, the specimens and test chamber were cooled to -25°C by liquid nitrogen and then heated. Immediately after the first run had been made on a given specimen, the specimen was cooled to -25°C and a second run was made. Data were analyzed statistically by a two-way analysis of variance⁵, and means were compared by Tukey's method⁶ at the 95 percent level of confidence.

Results.

Linear coefficients of thermal expansion (α) were calculated for three temperature ranges, 0 to 60°C, 10 to 45°C and 37°C. For the first two ranges, α was calculated by assuming $(\Delta l / \Delta T)$ in the aforementioned equation was a straight line between the two respective temperatures. The value of α at 37°C was calculated by equating the tangent of the thermal expansion curve at 37°C equal to $(\Delta l / \Delta T)$, where the tangent was determined graphically. Values of α determined for the aforementioned conditions are listed in Table 2 for runs 1 and 2 for the composites and sealants. Values of α for P were obtained at powder/liquid ratios of 1/1 and 1/2 as indicated in Table 2.

Two-way analyses of variance were made for each temperature range separately for the composites and for the sealants to compare the effects of products and runs on α .

TABLE 2
 LINEAR COEFFICIENTS OF THERMAL EXPANSION OF COMPOSITES AND SEALANTS FOR
 THREE TEMPERATURE RANGES.

MATERIAL	Linear Coefficient of Thermal Expansion*, $\times 10^{-6}/^{\circ}\text{C}$					
	0 to 60°C†		10 to 45°C‡		37°C§	
	Run 1	Run 2	Run 1	Run 2	Run 1	Run 2
Composites:						
A	40.1 (1.2)	35.8 (0.7)	40.0 (1.5)	34.2 (0.5)	49.8 (1.7)	39.4 (0.7)
AR	39.0 (1.0)	32.2 (2.5)	37.3 (1.1)	30.1 (3.0)	46.4 (1.1)	36.7 (1.8)
C	43.4 (1.4)	39.6 (2.2)	47.2 (2.0)	38.0 (2.3)	60.5 (2.0)	44.2 (2.0)
NF	30.2 (2.4)	28.3 (1.3)	28.2 (2.3)	27.5 (1.4)	31.7 (1.8)	30.6 (1.0)
P(1/1)	43.2 (2.0)	37.8 (2.1)	39.2 (1.8)	36.8 (1.9)	44.5 (2.6)	38.9 (2.6)
P(1/2)	53.0 (2.1)	47.3 (1.5)	51.6 (1.7)	46.2 (1.2)	60.9 (2.2)	51.7 (1.7)
S	31.6 (2.4)	28.2 (1.3)	31.6 (1.5)	26.6 (1.2)	41.6 (1.7)	32.0 (1.7)
V	27.8 (1.7)	26.5 (1.6)	27.0 (1.9)	24.9 (2.1)	35.1 (1.9)	30.3 (1.3)
Sealants:						
D	97.1 (1.8)	90.3 (1.2)	93.5 (1.6)	88.9 (1.4)	103.5 (2.0)	96.1 (1.2)
K	78.2 (4.2)	70.9 (3.9)	76.5 (4.7)	68.5 (3.6)	90.9 (6.1)	79.0 (4.7)
NS	80 (9)	78 (8)	78 (10)	78 (9)	86 (13)	85 (11)
W	99.1 (2.6)	93.7 (1.2)	96.4 (3.6)	92.5 (1.0)	108.9 (3.6)	103.5 (1.2)

*Mean value of five replications with standard deviations in parentheses.

†Tukey's intervals at the 95% level for comparisons among products and between runs were 2.5 and 0.8 for composites and 3.1 and 2.1 for sealants excluding NS.

‡Tukey's intervals at the 95% level for comparisons among products between runs were 2.5 and 0.8 for composites and 3.3 and 2.2 for sealants excluding NS.

§Tukey's intervals at the 95% level for comparisons among products and between runs were 2.6 and 0.8 for composites and 4.0 and 2.7 for sealants excluding NS.

In each analysis, the effects of products, runs, and their interaction were significant statistically at the 95 percent level of confidence. Tukey's intervals for comparisons among means are listed in the footnotes of Table 2. The data for NS were not included in the analyses because the variance was unusually high.

Reheating of samples immediately after cooling from the first run resulted in changes in the thermal expansion curves as shown in Figure 1 for P (1/1). Subsequent reheating between 0 to 60°C produced no further changes in the thermal behavior. Values of α determined between 0 and 60°C were lower for run 2 than run 1, the percent difference between them ($\alpha_{\text{run 2}} - \alpha_{\text{run 1}} / \alpha_{\text{run 1}}$) being: A, -10.7; AR, -17.4; C, -8.8; NF, -6.6; P(1/1), -12.5; P(1/2), -10.8; S, -10.8; and V, -4.7 among the composites and D, -7.0; K, -9.3; NS, -2.5; and W, -5.4 among the sealants. The increased dimensional change associated with run 1 occurred at temperatures between 40 and 60°C for the composites and sealants tested.

Among the composite resins tested, the glass-filled materials (NF, S and V) had

lower values of α (0 to 60°C, run 2), ranging from 26.5 to 28.2 $\times 10^{-6}/^{\circ}\text{C}$, than the materials containing primarily quartz filler (A, AR, C, and P) with values of α ranging from 32.2 to 43.7 $\times 10^{-6}/^{\circ}\text{C}$. Among the sealants tested, α (0 to 60°C, run 2) varied from 70.9 to 93.7 $\times 10^{-6}/^{\circ}\text{C}$. The quartz-filled sealant (K) had lower values of α than the unfilled products. As a group, the sealants had much larger values of α than the composite resins.

Discussion.

The additional dimensional change associated with the thermal expansion curve of run 1 compared to run 2 between 40 and 60°C probably is caused by a release of residual compressive stress that is incorporated in the material during polymerization. The temperature at which this apparent release of stress begins is probably the glass transition temperature of the polymer phase of the materials tested.

Clinically, it is desirable for the thermal dimensional changes of restorative materials to approximate those of tooth structure to control marginal leakage and to maintain enamel bonding. The thermal coefficients of

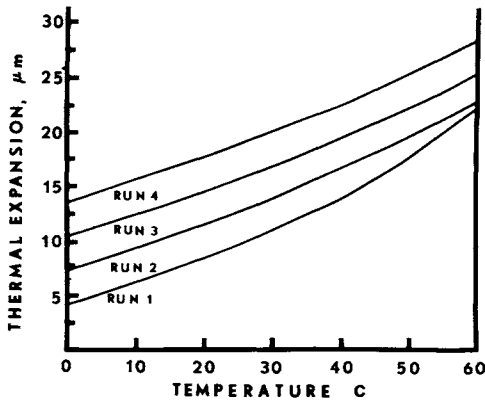


Fig. 1. -- Thermal expansion curves for P(1/1) showing differences in the dimensional change between the first run and subsequent runs. Curves are offset from each other for convenience.

expansion of the composites, and particularly the sealants, continue to be well above that of human tooth structure (about $11.4 \times 10^{-6}/^{\circ}\text{C}$).⁷ The true clinical picture is quite complicated because thermal equilibrium is seldom obtained in the oral environment. The thermal diffusivity of composite resins⁸ ($0.675 \text{ mm}^2/\text{sec}$) is greater than the values reported for either enamel ($0.469 \text{ mm}^2/\text{sec}$) or dentin⁹ ($0.183 \text{ mm}^2/\text{sec}$). A thin coating of resin, as is utilized for sealing occlusal pits and fissures, has a greater potential for reaching thermal equilibrium than the bulk of material found in a restoration. The thinness of a coating does, however, help to minimize the actual dimensional change.

The effect of decreasing filler content to reduce viscosity in order to gain enamel bonding from a single mix of material can be observed by comparing P(1/1) and P(1/2). In decreasing the inorganic filler content 5 percent from 70.4 percent in P(1/1) to 65.8 percent in P(1/2)¹⁰, the linear coefficient of thermal expansion increased 25 percent from 37.8 to $47.3 \times 10^{-6}/^{\circ}\text{C}$ between 0 and 60°C . There is, therefore, some indication that the thermal dimensional change should be less when a bonding agent is used with a viscous composite resin rather than a single mix resin-system with lower viscosity and decreased filler content.

Conclusions.

Linear thermal coefficients of expansion (α) of seven commercial composite restorative resins and four pit and fissure sealants were determined between 0 and 60°C on a thermomechanical analyzer. The thermal expansion curves obtained were nonlinear over this temperature range. Values of α determined upon reheating samples immediately after cooling from the first run decreased by as much as 17 percent. Upon reheating, values of α ranged from 26.5 to $39.9 \times 10^{-6}/^{\circ}\text{C}$ from the composites and from 70.9 to $93.7 \times 10^{-6}/^{\circ}\text{C}$ for the pit and fissure sealants between 0 and 60°C . Composites with glass filler had lower values of α than ones with quartz filler.

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