

# Polymerization Behavior and Mechanical Properties of High-Viscosity Bulk Fill and Low Shrinkage Resin Composites

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## Clinical Relevance

Bulk fill resin composites may be less suitable than low-shrinkage resin composites for the restoration of large cavities due to their high volumetric shrinkage.

## SUMMARY

The present study determined the mechanical properties and volumetric polymerization shrinkage of different categories of resin composite. Three high viscosity bulk fill resin composites were tested: Tetric EvoCeram Bulk Fill (TB, Ivoclar Vivadent), Filtek Bulk Fill posterior restorative (FB, 3M ESPE), and Sonic Fill (SF, Kerr Corp). Two low-shrinkage resin composites, Kalore (KL, GC Corp) and Filtek

LS Posterior (LS, 3M ESPE), were used. Three conventional resin composites, Herculite Ultra (HU, Kerr Corp), Estelite Quick (EQ, Tokuyama Dental), and Filtek Supreme Ultra (SU, 3M ESPE), were used as comparison materials. Following ISO Specification 4049, six specimens for each resin composite were used to determine flexural strength, elastic modulus, and resilience. Volumetric polymerization shrinkage was determined using a water-filled dilatometer. Data were evaluated using analysis of variance followed by Tukey's

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honestly significant difference test ( $\alpha=0.05$ ). The flexural strength of the resin composites ranged from 115.4 to 148.1 MPa, the elastic modulus ranged from 5.6 to 13.4 GPa, and the resilience ranged from 0.70 to 1.0 MJ/m<sup>3</sup>. There were significant differences in flexural properties between the materials but no clear outliers. Volumetric changes as a function of time over a duration of 180 seconds depended on the type of resin composite. However, for all the resin composites, apart from LS, volumetric shrinkage began soon after the start of light irradiation, and a rapid decrease in volume during light irradiation followed by a slower decrease was observed. The low shrinkage resin composites KL and LS showed significantly lower volumetric shrinkage than the other tested materials at the measuring point of 180 seconds. In contrast, the three bulk fill resin composites showed higher volumetric change than the other resin composites. The findings from this study provide clinicians with valuable information regarding the mechanical properties and polymerization kinetics of these categories of current resin composite.

## INTRODUCTION

The mechanical properties of resin composites have improved following the development of superior filler components and resin monomers.<sup>1</sup> However, one of the major shortcomings of resin composites is the shrinkage that accompanies polymerization.<sup>2,3</sup> In addition, the viscosity of the resin composites increases during the polymerization process, leading to increased stiffness of the material.<sup>4,5</sup> As a result, contraction stress occurs not only within the material but also at the bonded interface.<sup>3</sup> It is thought that this morphologic alteration leads to negative sequelae, such as postoperative sensitivity, contraction gaps, microcracks in the tooth structure, cuspal deformation, and recurrent caries.<sup>6-8</sup> To reduce the effects of polymerization shrinkage and contraction stress, various clinical approaches have been attempted. For instance, a sandwich technique with glass-ionomer cements,<sup>9</sup> incremental filling techniques,<sup>10,11</sup> and the use of low-viscosity flowable resin composites<sup>12,13</sup> have been advocated. However, these techniques increase the number of clinical steps and may increase technique sensitivity.

Polymerization of light-cured resin composites is influenced by a wide range of factors, including shade, light absorption and dispersion within the

resin composite, types of fillers and monomer, and light irradiance conditions.<sup>14</sup> In particular, resin monomers have a decisive influence on the characteristics of polymerization.<sup>15,16</sup> In general, during the course of polymerization, the intermolecular separations between the dimethacrylate monomers are reduced due to the conversion of carbon-carbon double (C=C) bonds, resulting in volumetric shrinkage.<sup>2,17</sup> Hence, over recent decades, research and development have focused on creating and adopting new resin monomers to reduce polymerization shrinkage and contraction stress. Recently, new resin monomers with increased molecular weight and a small number of C=C bonds<sup>18,19</sup> or cationic polymerization<sup>20</sup> have been introduced in several low-shrinkage resin composite products.<sup>21</sup>

It should be noted that other trends in resin composite development are also of interest. To reduce contraction stress and the number of clinical steps, bulk fill resin composites have recently been marketed. Bulk fill resin composites are intended to allow placement in one layer of up to 4 mm thickness with adequate polymerization.<sup>22-24</sup> In addition, these composites have a short activation time due to the presence of modified initiation systems<sup>25</sup> and increased translucency owing to decreased filler load and increased filler size.<sup>26</sup> Although there is no consensus on the classification, bulk fill resin composites are described either as low-viscosity that are similar to flowable resin composites or as high-viscosity that resemble conventional resin composites.<sup>27</sup> However, *in vitro*-simulated wear studies showed that bulk fill resin composites have lower wear resistance compared with conventional resin composites.<sup>28</sup> Therefore, independent evaluations of the materials are important if dental professionals are to make an informed choice of the optimal product.

There have been many studies investigating the mechanical properties and polymerization kinetics of low-shrinkage<sup>18,19,21,29,30</sup> and bulk fill resin composites.<sup>22-27,31,32</sup> However, few studies have compared the mechanical properties and polymerization behaviors of different types of resin composite under the same conditions. The purpose of this study was to assess the mechanical properties and volumetric polymerization shrinkage of currently available resin composites, including bulk fill and low-shrinkage resin composites, and to compare their characteristics with those of the conventional resin composites. The null hypothesis to be tested was that there are no significant differences in mechan-

Table 1: *Materials used in this study*

Code	Resin composite (shade: lot no.)	Main components	Manufacturer
TB	Tetric EvoCream Bulk Fill (Shade; A2: T21387)	bis-GMA, UDMA, ytterbium trifluoride, Ba glass filler, mixed oxide, prepolymer filler, EBPADMA, additives, catalysts, stabilizers, pigments	Ivoclar vivadent
FB	Filtek Bulk Fill posterior restorative (Shade; A2: N701975)	silane treated creamic, aromatic UDMA, ytterbium fluoride, UDMA, silane treated silica, DDDMA, silane treated zirconia, water, modified methacrylate monomer, EDMAB benzotriazol	3M ESPE
SF	SonicFill (Shade; A2:5037026)	poly(oxy-1,2-ethanediyl), $\alpha$ , $\alpha'$ -[(1-methylethylidene) $\beta$ di-4, 1-phenylene]bis [ $\omega$ -[(2-methyl-1-oxo-2-propen-1-yl)oxy]-2,2'-ethylenedioxydiethyl dimethacrylate, bis-GMA, TEGDMA, bis-EMA, barium glass, silica, rheological modifier	Kerr Corp
KL	Kalore (Shade; A2: 9EP)	ytterbium trifluoride, UDMA, bis-EMA, DX-511, dimethacrylate, BHT, silane	GC Corp
LS	Filtek LS (Shade; A2: 9EP)	silane treated quartz, ytterbium trifluoride 3,4-epoxycyclohexylcyclopolydimethylsiloxane, Bis-3,4-epoxycyclohexylethyl-phenyl-methylsilane, Borate (1-), thtrakis (pentafluorophenyl) -[4-(methylethyl) phenyl] (4-methylphenyl) iodonium	3M ESPE
HU	Herculite Ultra (Shade; EA2: 4495606)	bis-GMA, TEGDMA, bis-EMA barium glass, silica, prepolymerized filler	Kerr Corp
EQ	Estelite $\Sigma$ Quick (Shade; A2:154095P)	bis-GMA, TEGDMA, silica-zirconia filler, dibutyl hydroxy toluene	Tokuyama Dental
SU	Filtek Supreme Ultra (Shade; BA2: N339152)	bis-GMA, bis-EMA, UDMA, TEGDMA, PEGDMA, silica nanofiller, zirconia/silica Aggregated zirconia/silica clusters	3M ESPE

Abbreviations: BHT, butylated hydroxytoluene; bis-EMA, bisphenol A polyethethylene glycol diether dimethacrylate; bis-GMA, 2, 2-bis[4-(2-hydroxy-3-methacryloyloxypropoxy)phenyl]propane, DDDMA, 1,12-dodecane dimethacrylate; EBPADMA, ethoxylated bisphenol A dimethacrylate; EDMAB, ethyl 4-dimethyl aminobenzoate; PEGDMA, poly ethethylene glycol diether dimethacrylate; TEGDMA, triethylene glycol dimethacrylate; UDMA: urethane dimethacrylate. A Sonic Fill Handpiece 2010 (lot no. 1007885) was used for insertion of the SonicFill material.

ical properties and polymerization behaviors among the three different types of resin composites.

## METHODS AND MATERIALS

Three high-viscosity bulk fill resin composites were tested: Tetric EvoCeram Bulk Fill (TB, Ivoclar Vivadent, Schaan, Liechtenstein), Filtek Bulk Fill posterior restorative (FB, 3M ESPE, St Paul, MN, USA), and SonicFill (SF, Kerr Corp, Orange, CA, USA). Two low-shrinkage resin composites, Kalore (KL, GC Corp, Tokyo, Japan) and Filtek LS Posterior (LS, 3M ESPE), were used. Three conventional resin composites, Herculite Ultra (HU, Kerr Corp), Estelite  $\Sigma$  Quick (EQ, Tokuyama Dental, Tokyo, Japan), and Filtek Supreme Ultra (SU, 3M ESPE), were used as comparison materials. The test materials and their components are listed in Table 1. To avoid any influence from the reported nonuniformity of LED curing units,<sup>33,34</sup> a quartz-tungsten-halogen curing unit was used (Optilux 501, sds Kerr, Danbury, CT, USA) with a curved light guide (type 20812; diameter, 13 mm; sds Kerr), and the light irradiance (average 600 mW/cm<sup>2</sup>) of the curing unit was checked using a dental radiometer (Model 100, Kerr).

## Inorganic Filler Content

The inorganic filler content of the materials was measured using thermogravimetry and differential thermal analysis (TG/DTA6300, Seiko Instruments, Tokyo, Japan). For each resin composite tested, resin paste (50 mg) was placed in a cylindroid crucible of pure platinum (7 mm in diameter, 10 mm in depth) and heated in the thermogravimeter from 25°C to 800°C at a heating rate of 10°C/min under atmospheric air until the organic components were completely incinerated. The weight of the residual resin paste was automatically measured by the built-in horizontal differential balance, and the inorganic filler content (wt%) was calculated using the compensated blank curve. Five measurements per test material were conducted to obtain an average inorganic filler content (wt%).

## Coefficient of Linear Thermal Expansion

The coefficients of linear thermal expansion of the test materials were measured using a thermomechanical analyzer (TMA/SS 6300, Seiko Instruments). For each resin composite tested, the resin paste was condensed into a cylindrical Teflon (Sanplatec Corp, Osaka, Japan) mold (8.0 mm in

high, 3.0 mm in diameter). Light irradiance was performed at both the top and bottom of the mold for 40 seconds; the mold was then cut with a sharp scalpel and removed from the cured resin composite. To ensure full polymerization, the center of the specimen was irradiated perpendicular to the axis for 40 seconds; this was then repeated on the opposite side of the specimen. The specimens were stored under dark conditions at 25°C for 24 hours before the measurements were conducted. Each specimen was placed on the stage of the chamber, and the thermomechanical analyser probe contacted the top surface of the specimen at 50-mN load stress. Five specimens were prepared for each material and separately tested at a heating rate of 2°C/min from 25°C to 130°C under atmospheric air. Measurements were performed four times per specimen, but the first measurement outcome was discarded as unreliable. The average coefficient of linear thermal expansion ( $\times 10^{-6}/^{\circ}\text{C}$ ) was measured over a temperature range of 50°C (from 30°C to 80°C), the same range width as that used in standard thermal cycling experiments, starting at a low temperature of 30°C to avoid any influence of fluctuations in ambient temperature.

### Flexural Properties

Flexural properties of the resin composites were tested according to the International Standards Organization Standard 4049.<sup>35</sup> Each resin composite was compacted into a stainless-steel split mold with dimensions  $25 \times 2 \times 2$  mm and positioned on a glass slide. The middle third of the specimen was first irradiated for 30 seconds, after which the remaining thirds were irradiated for 30 seconds each. The opposite side of the specimen was irradiated in the same manner. After the removal of the hardened specimen from the mold, all six sides were wet polished with #1200 silicon carbide (SiC) paper (Fuji Star Type DDC, Sankyo Rikagaku Co, Ltd, Saitama, Japan). The specimens were then stored for 24 hours in distilled water at 37°C before conducting the mechanical tests. Six specimens per test group were subjected to a three-point bending flexural strength test using a universal testing instrument (Type 5500R, Instron Corp, Canton, MA, USA) at a cross-head speed of 1.0 mm/min until the specimen fractured. The specimens were positioned on a three-point bending apparatus with a span length of 20.0 mm. Flexural strength (S) and modulus of elasticity (E) were determined from the stress-strain curve obtained using the Bluehill Ver 2.5 computer software (Instron Corp) linked to the testing instru-

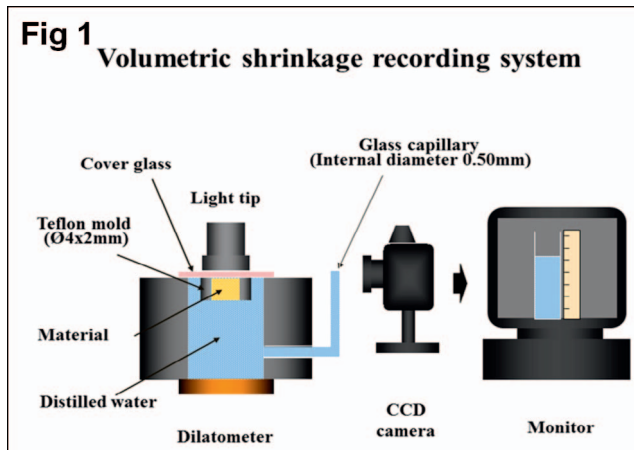


Figure 1. Diagram of the volumetric polymerization shrinkage recording system.

ment. The modulus of resilience (R) was calculated using the following equation by Peutzfeldt & Asmussen<sup>36</sup>:  $R = S^2/2E$ .

### Volumetric Shrinkage Measurement

For volumetric polymerization shrinkage measurements, the test apparatus was composed of a water-filled dilatometer and a capillary tube (uniform diameter, 0.5 mm; length, approximately 130 mm). It was attached to a 25 cm<sup>3</sup>, brass-bottomed density bottle using a ground glass joint. A diagram of the volumetric shrinkage recording system is shown in Figure 1. The density bottle was filled with distilled water. During the course of testing, the temperature of the distilled water was maintained by placing the density bottle on a thermostatically controlled plate to avoid the influence of temperature changes in the water.

Resin composite paste was placed into a Teflon mold (4.0 mm diameter  $\times$  2.0 mm height) and covered by a 0.5-mm-thick glass plate. The curing light tip end was placed in a hole on the glass lid, and the specimen was light irradiated for 30 seconds. The change in the height of the water meniscus was recorded using a CCD camera (DS-505, Nikon Corp, Tokyo, Japan) every five seconds from light irradiation to 180 seconds and projected onto a VCR (CT-1450, Hitachi Corp, Tokyo, Japan) from the start of light irradiation. A period of 180 seconds was chosen because polymerization shrinkage tends to plateau after about 120 seconds, and water absorption affects volume over longer periods. Measurement accuracy of the water meniscus was 0.2 mm.

Volumetric shrinkage ( $\Delta V$ ) of the specimens was calculated from the change in the height of water

Table 2: Inorganic filler contents and thermal expansion

Code	Inorganic filler content (wt%)	Tukey group	Coefficient of linear thermal expansion	Tukey group
TB	77.1 (0.4)	b	42.2 (0.8)	c
FB	75.1 (0.5)	c	48.9 (0.7)	a
SF	81.3 (0.5)	a	46.1 (0.4)	b
KL	69.0 (0.4)	e	36.6 (0.6)	e
LS	77.0 (0.4)	b	33.5 (0.9)	f
HU	67.2 (0.3)	f	39.0 (1.1)	d
EQ	67.6 (0.4)	f	41.8 (0.6)	c
SU	73.7 (0.5)	d	38.0 (0.9)	d

Values are mean (SD). Same lowercase letter in vertical columns indicates no difference at 5% significance level.

meniscus ( $\Delta h$ ) using the equation  $\Delta V = 0.25\pi \times \Delta h \times d^2$ , where  $d$  is the diameter of the capillary tube. The percentage volumetric change was then calculated. Five specimens per material were measured, and the average volumetric shrinkage was obtained.

### Scanning Electron Microscopy Observations

The surfaces of the cured resin composite specimens were polished to a high gloss with abrasive discs (Fuji Star Type DDC, Sankyo Rikagaku Co) and a series of diamond pastes down to 0.25- $\mu$ m particle size (DP-Paste, Struers, Ballerup, Denmark). The polished surfaces were then subjected to unfiltered argon ion beam etching (IIS-200ER, Elionix, Tokyo, Japan) for 45 seconds in the direction perpendicular to the polished surface at an accelerating voltage of 1.0 kV and ion current density of 0.4 mA/cm<sup>2</sup>. This is known to improve the visibility of the layers in the sample.<sup>37</sup> The surfaces were coated in a vacuum evaporator (Quick Coater Type SC-701, Sanyu Denshi Inc, Tokyo, Japan) with a thin film of gold (thickness count, 300). Examinations of the surfaces were carried out by scanning electron microscopy (SEM; FE-8000, Elionix) with an operating voltage of 10 kV and at magnifications of 5000 $\times$  and 20,000 $\times$ .

### Statistical Analysis

Because of their homogeneity of variance (Bartlett's test) and normal distribution (Kolmogorov-Smirnov test), the data for each material were subjected to analysis of variance followed by Tukey's honestly significant difference test at a significance level of 0.05. Bartlett's test was performed with a custom program implemented in a spreadsheet (Excel, Microsoft Inc, Redmond, WA, USA), and the other statistical analysis was conducted using a statistical software system (Sigma Plot ver. 11.0, SPSS Inc, Chicago, IL, USA).

## RESULTS

### Inorganic Filler Content and Coefficient of Thermal Expansion

The inorganic filler contents and coefficients of linear thermal expansion are shown in Table 2. The average inorganic filler contents of the resin composites ranged from 67.2 to 81.3 wt%. The resin composites used in this study had significantly different inorganic filler contents, and the bulk-fill resin composite SF had the highest value. The bulk fill resin composites tended to have higher inorganic filler contents than the other resin composites.

The average coefficients of thermal expansion of the resin composites ranged from 33.5 to 48.9 ( $\times 10^{-6}/^{\circ}\text{C}$ ). Although the bulk fill resin composites exhibited higher thermal expansion, the low-shrinkage resin composites had lower thermal expansion than the conventional resin composites.

### Flexural Properties

The results of flexural properties testing are shown in Table 3. The flexural strength of the resin composites ranged from 116.9 to 148.1 MPa, the elastic modulus ranged from 5.6 to 13.4 GPa, and average resilience ranged from 0.70 to 1.0 MJ/mm<sup>3</sup>. There were significant differences between the

Table 3: Flexural strength, elastic modulus, and resilience<sup>a</sup>

Code	Flexural strength (MPa)	Tukey group	Elastic modulus (GPa)	Tukey group	Resilience (MJ/mm <sup>3</sup> )	Tukey group
TB	115.4 (5.0)	d	9.0 (0.8)	d	0.74 (0.15)	a,b
FB	138.1 (7.5)	a,b	10.5 (0.9)	c,d	0.92 (0.11)	a,b
SF	120.0 (6.5)	c,d	5.6 (0.5)	e	0.78 (0.15)	a,b
KL	116.9 (7.3)	c,d	8.9 (0.4)	d	0.77 (0.20)	a,b
LS	129.7 (7.9)	b,c	12.1 (0.5)	a,b	0.70 (0.15)	b
HU	148.1 (7.0)	a	11.0 (1.3)	b,c	1.0 (0.15)	a
EQ	120.5 (8.5)	c,d	9.6 (0.6)	d	0.75 (0.14)	a,b
SU	138.2 (7.2)	a,b	13.4 (0.5)	a	0.71 (0.13)	b

<sup>a</sup> Values are mean (SD). Same lowercase letter in vertical columns indicates no difference at 5% significance level.

Table 4: Volumetric change of resin composites as a function of time <sup>a</sup>					
Code	10 seconds	30 seconds	60 seconds	120 seconds	180 seconds
TB	−0.68 (0.05)a,A	−1.75 (0.05)a,B	−1.94 (0.05)b,C	−2.19 (0.10)b,D	−2.19 (0.09)b,D
FB	−0.49 (0.06)b,A	−1.37 (0.06)b,B	−1.62 (0.08)c,C	−1.93 (0.06)c,D	−1.95 (0.08)c,D
SF	−0.59 (0.06)ab,A	−1.37 (0.07)b,B	−2.15 (0.09)a,C	−2.91 (0.12)a,D	−2.93 (0.13)a,D
KL	−0.48 (0.05)b,A	−0.66 (0.06)d,B	−0.88 (0.06)g,C	−1.05 (0.08)f,D	−1.11 (0.08)g,D
LS	0.21 (0.05)d,A	−0.18 (0.06)e,B	−0.39 (0.05)h,C	−0.83 (0.06)g,D	−1.02 (0.06)g,E
HU	−0.35 (0.06)c,A	−0.96 (0.08)c,B	−1.39 (0.07)d,C	−1.56 (0.10)d,D	−1.56 (0.06)e,D
EQ	−0.57 (0.07)b,A	−0.78 (0.09)d,B	−1.05 (0.06)f,C	−1.37 (0.05)e,D	−1.37 (0.10)f,D
SU	−0.49(0.08)b,A	−0.76(0.06)d,B	−1.2 (0.09)e,C	−1.58 (0.06)d,D	−1.67 (0.09)d,D

<sup>a</sup> Values are mean (SD). Same lowercase letter in vertical columns indicates no difference at 5% significance level. Same uppercase letter in horizontal columns indicates no difference at 5% significance level.

flexural strengths but no clear outliers. Regarding the elastic modulus, the bulk fill SF showed a significantly lower value than the other resin composites. HU showed the highest resilience, but it was significantly different only from LS and SU.

Polymerization Shrinkage

The results for volumetric changes are shown in Table 4 and Figure 2. Volumetric changes as a function of time over a duration of 180 seconds depended on the type of resin composite. However, for all the resin composites, apart from LS, volumetric shrinkage began soon after the start of light irradiation, and a rapid decrease in volume during light irradiation followed by a gradual decrease was observed (Figure 2). For the measuring point at 10 seconds, volumetric changes ranged from +0.21% to −0.68%. Only LS showed a volumetric increase, and the difference between LS and the other resin

composites was statistically significant. For the measuring point at 180 seconds, volumetric changes ranged from −1.02% to −2.93%. The highest volumetric shrinkage was seen with SF, whereas LS showed the lowest final shrinkage among the tested materials. No significant differences in final shrinkage were observed in the low-shrinkage resin composites KL and LS. On the other hand, the bulk fill resin composites TB, FB, and SF showed higher volumetric changes compared with the other resin composites. Based on the volumetric changes, the resins were segregated into three groups: KL and LS belonged to the lower shrinkage group; HU, EQ, and SU formed the middle shrinkage group; and TB, FB, and SF were in the higher shrinkage group. All the tested materials continued to exhibit slower volumetric changes even after cessation of the light irradiation. Although there was a significant difference between the measuring point at 60 seconds and

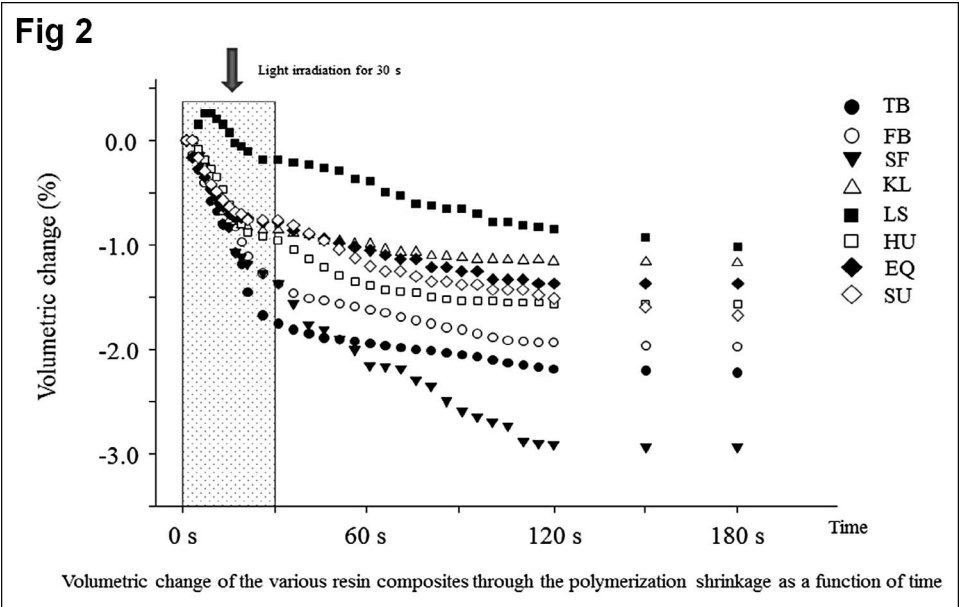


Figure 2. Volumetric changes in the various resin composites due to polymerization shrinkage as a function of time.



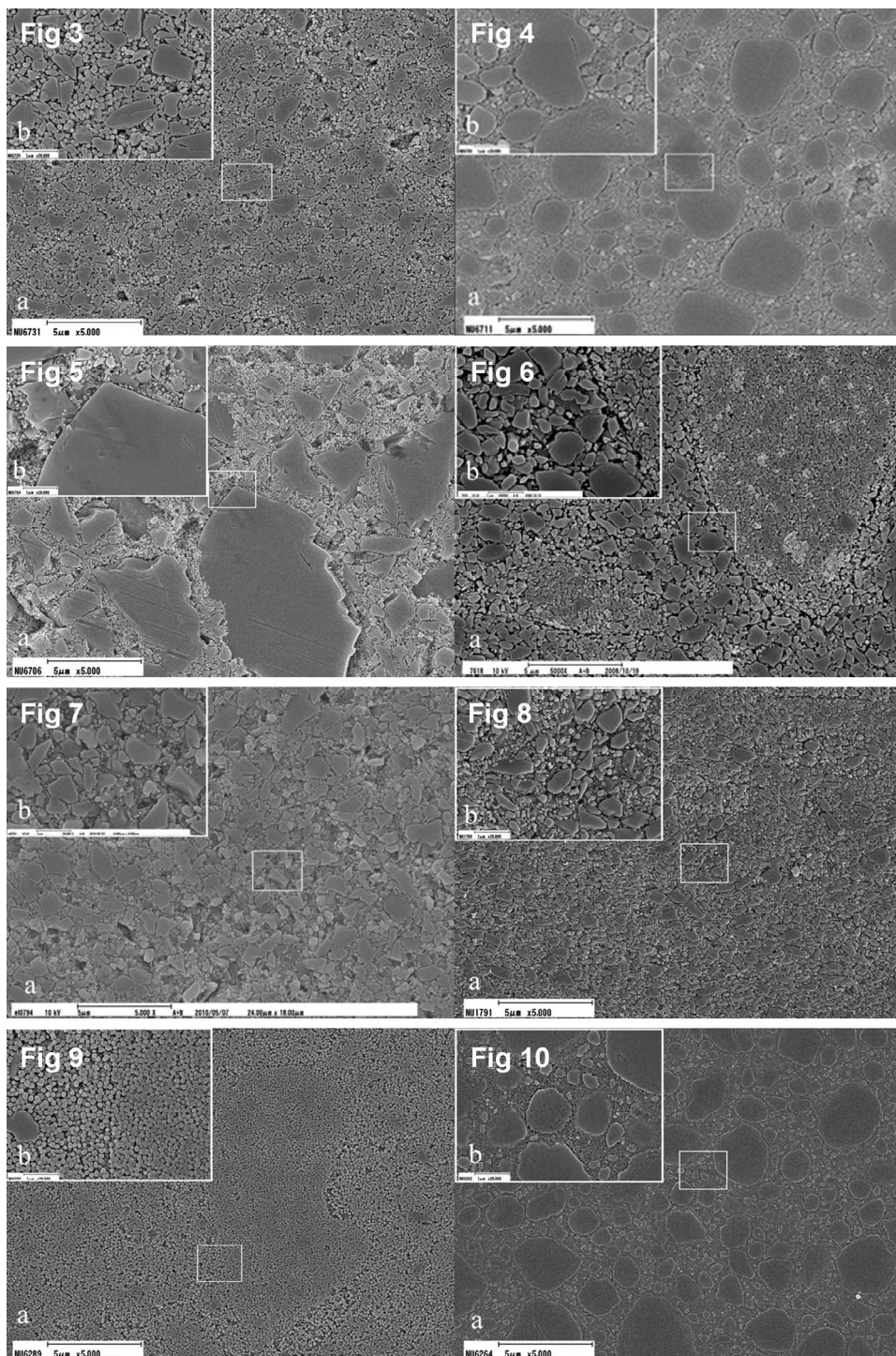


Figure 3. *Tetric EvoCeram Bulk Fill*—argon ion-etched surface (a) 5000 $\times$  and (b) 20,000 $\times$ .

Figure 4. *Filtek Bulk Fill posterior restorative*—argon ion-etched surface (a) 5000 $\times$  and (b) 20,000 $\times$ .

Figure 5. *Sonic Fill*—argon ion-etched surface (a) 5000 $\times$  and (b) 20,000 $\times$ .

Figure 6. *Kalore*—argon ion-etched surface (a) 5000 $\times$  and (b) 20,000 $\times$ .

Figure 7. *Filtek LS*—argon ion-etched surface (a) 5000 $\times$  and (b) 20,000 $\times$ .

Figure 8. *Herculite Ultra*—argon ion-etched surface (a) 5000 $\times$  and (b) 20,000 $\times$ .

Figure 9. *Estelite Sumi Quick*—argon ion-etched surface (a) 5000 $\times$  and (b) 20,000 $\times$ .

Figure 10. *Filtek Supreme Ultra*—argon ion-etched surface (a) 5000 $\times$  and (b) 20,000 $\times$ .

that at 120 seconds, no significant differences were noted at 120 and 180 seconds for all the materials, apart from LS.

### SEM Observations

SEM images of the eight resin composites after argon ion etching are presented in Figures 3–10.

These figures illustrate the differences in filler shape, particle size and distribution. TB, LS, and HU had similar appearances with 0.2- to 2- $\mu$ m milled irregularly shaped glass filler particles, as shown in Figures 3, 7, and 8, respectively. FB and SU had rounded nano-sized zirconia/silica filler particles and 0.5- to 5- $\mu$ m spheroidal aggregates of nano-sized

fillers (Figures 4 and 10). EQ also had spherical nano-sized filler particles, but 5- to 10- $\mu\text{m}$  irregular aggregated fillers were observed (Figure 9). The glass filler particles of SF (Figure 5) were larger than the fillers in the other resin composite systems containing ground glass filler particles (TB, LS, and HU). KL had 0.2- to 1- $\mu\text{m}$  irregularly shaped glass filler particles and 5- to 20- $\mu\text{m}$  irregular aggregates of glass filler (Figure 6).

## DISCUSSION

The aim of this study was to aid the clinical selection of resin composites by measuring the mechanical properties and polymerization behavior of resin composites belonging to various categories. Clear differences between the tested high-viscosity bulk fill resin composites and the other types of resin composite were shown.

For bulk fill resin composites, several developments and technologies have contributed to a deeper cure. To enhance the matrix resin degree of conversion, new initiator systems with a higher photocuring activity than camphorquinone are used,<sup>25</sup> or the translucency of the resin is adjusted to increase the transmission of light.<sup>2,38</sup> In addition, larger filler particles are used to reduce light scattering. Although initial low-viscosity bulk fill resin composites seemed to have good cavity wall adaptation due to flowability, they did not have adequate surface and bulk properties for endurance in high-stress bearing areas.<sup>39</sup> The tested high-viscosity bulk fill resin composites showed slightly higher inorganic filler content than the other resin composites. Improved handling, surface, and bulk mechanical properties, compared with the low-viscosity bulk fill resin composites, have been attributed to the higher filler content.

A large difference in thermal expansion induces gaps in the vicinity of the resin/tooth interface, resulting in the penetration of oral fluids between the restoration and the tooth surface.<sup>40</sup> This could lead to irritation of the dental pulp, marginal degradation, and secondary decay. A higher inorganic filler content is expected to reduce thermal expansion because of the reduction in the amount of resin matrix, which is more sensitive to thermal changes. However, despite the higher inorganic filler content in the bulk fill resin composites, they exhibited significantly higher thermal expansion compared with the other resin composites in this study. It has been reported that no correlation exists between the inorganic filler content and the coefficient of thermal expansion of the resin cements and

dual-cured provisional resins.<sup>41,42</sup> Therefore, regardless of the type of resin composite, other factors such as the type of matrix resin, molecular mobility, surface treatment of fillers, and size of filler particles might have a significant impact on the thermal properties.

Despite having the highest inorganic filler content among the bulk-fill resin composites, SF had a relatively low flexural strength and a significantly lower elastic modulus compared with the other resin composites. Previous research has shown that SF showed significantly lower wear resistance compared with other bulk fill and conventional resin composites, and SEM observations revealed that plucking of filler particles was obvious after wear testing.<sup>28</sup> This finding is consistent with our results that SF had a lower flexural strength and elastic modulus compared with the other resin composites because flexural properties and wear resistance are related.<sup>1</sup> In particular, it is possible that the large glass filler particles in SF (Figure 5) create easy routes for crack propagation between the glass fillers and resin matrix.

All the tested bulk fill resin composites showed significantly higher volumetric shrinkage than the other resin composite types. In particular, SF showed approximately twice the volumetric shrinkage as the conventional composites HU and EQ. To obtain good adaptation to the cavity wall, SF uses a diluted triethylene glycol dimethacrylate monomer and a rheological modifier to support the enhancement of the flowability of the resin paste with ultrasound.<sup>43</sup> It can be speculated that, although these components of the resin matrix may contribute to increased volumetric shrinkage, they, along with lower elastic modulus, may also cause a reduction in polymerization shrinkage stress. The viscoelastic behavior of a resin-based material, as determined by their flow during the early stages of polymerization and the increase in elastic modulus, plays an important role in polymerization contraction stress.<sup>16</sup> It has been reported that although low-viscosity bulk fill resin composites showed higher volumetric polymerization shrinkage than their high-viscosity counterparts, the contraction stresses of low-viscosity bulk fill resin composites were lower than those of the high-viscosity bulk fill resin composites.<sup>23</sup> Viscoelasticity and volumetric shrinkage are both determined by similar factors that make it difficult to separate their influence on the development of contraction stress.<sup>12,44</sup> The complexity of contraction stress means that it is important to have a broad understanding of the properties of resin



composites when making clinical recommendations or decisions.<sup>16</sup>

For all the resin composites used in this study apart from LS, the volumetric shrinkage began soon after the start of light irradiation and polymerization shrinkage continued after the end of light irradiation. The shrinkage noted after removal of the light source might be attributed to the postirradiation reaction of residual monomers.<sup>45,46</sup> The low-shrinkage resin composites, KL and LS, showed significantly lower volumetric shrinkage compared with the other resin composites at measuring point 180 seconds. KL has prepolymerized filler particles, which include a lot of closely aggregated small splintered particles (Figure 6); it uses a new high-molecular-weight monomer rather than 2, 2-bis[4-(2-hydroxy-3-methacryloyloxypropoxy)phenyl]propane or urethane dimethacrylate.<sup>2,17</sup> The degree of shrinkage is dictated by the number of covalent bonds formed, which is the extent of the polymerization reaction.<sup>2,17</sup> The monomer in KL (DX-511) has a high molecular weight of 895 g/mol and a low number of carbon-carbon double bonds, which help to reduce polymerization shrinkage.<sup>18</sup> In contrast, LS adopts ring-opening chemistry in siloranes to replace methacrylates in the resin matrix.<sup>18,20,21</sup> Uniquely, LS showed volumetric increase during the first 10 seconds of light irradiation. This phenomenon might be attributed to the molecular structure of the silorane monomer and the polymerization process. During the initial phase of polymerization, intermolecular space may be expanded due to cationic ring opening polymerization of the cycloaliphatic oxirane moieties. Furthermore, the low-shrinkage resin composites KL and LS showed significantly lower thermal expansion than the other resin composites, and it could be speculated that even after the polymerization process is complete, the molecular mobility of the resin matrix in the low-shrinkage resin composites may be less than that of the other types of resin composite.

FB and SU, which were obtained from the same manufacturer, showed significantly higher flexural strength than most of the other tested materials. Although the two products have different intended uses and utilize different resin matrix monomers, the appearance and distribution of the fillers were found to be similar under SEM observation (Figures 4 and 10). Fillers can distribute the load stress and inhibit crack propagation due to pinning effects.<sup>47</sup> Therefore, for FB and SU, the balance of filler size, distribution of fillers, and the interaction between fillers and the resin matrix through silane coupling

may contribute effectively to dispersion of load stress. However, the elastic modulus and resilience of FB were different from those of SU, which may be attributed to a difference in resin monomers. For instance, only the high-viscosity bulk fill resin composite FB contains a 1,12-dodecane dimethacrylate (DDDMA) which has a hydrophobic backbone and increased molecular mobility. In addition, DDDMA provides flexibility, fast cure, and improved surface characteristics to the polymer matrix, which are suitable properties for bulk fill resin composites.<sup>48,49</sup> Furthermore, different resin monomers may influence the polymerization kinetics and thermal expansion of FB and SU, which were found to be different between the two resin types.

Lower elastic modulus may allow increased deformation and dimensional changes on the occlusal surfaces under high stress that could result in defect formation, marginal breakdown, or reduction of wear resistance.<sup>1</sup> In general, materials with high flexural strength also have a high elastic modulus, and these materials broadly follow that trend. However, LS has a high elastic modulus relative to its flexural strength. The elastic modulus of LS showed significantly higher values than the other resin composites, apart from SU and HU, whereas its flexural strength was only significantly higher than that of TB. LS uses an epoxy-based silorane system that involves the opening of an oxirane ring.<sup>20</sup> This epoxy-based silorane system contributes not only to the reduction of polymerization shrinkage but also to the evasion of the creation of an oxygen inhibited layer.<sup>18,19</sup> Thus, the resin monomer and resin have completely different properties from those of the other materials. Resilience is defined as the ability of a material to absorb energy when it is deformed elastically from external stress before failure.<sup>41</sup> Therefore, a high resilience is preferred for the long-term stability of restorations.<sup>50</sup> HU and FB showed somewhat higher resilience than the other resin composites in this study; however, the differences are not striking and their clinical effect is unclear.

The null hypothesis that there would be no significant differences in mechanical properties and polymerization behaviors among the three different categories of resin composite was rejected. In addition, even within the same category of resin composites, properties and state of fillers were material dependent. Therefore, it is important to understand the characteristics of each resin composite before clinical use and select the optimal one or combination in light of cavity size, depth, C-factors, and other factors. Furthermore, although mechanical properties

and polymerization behavior are not the only parameters for consideration in the selection of the optimal resin composite, these characteristics can provide valuable information and support the research and development of new resin composites.

### CONCLUSION

The results of this study provide useful additional information on the mechanical properties and polymerization behavior of different categories of resin composite. The tested bulk fill resin composites tended to have higher inorganic filler content and linear thermal expansion than the other categories. In contrast, low-shrinkage resin composites showed significantly lower thermal expansion than the other resin composites. The flexural properties of the tested resin composites were material dependent with no clear outliers. The low-shrinkage resin composites showed significantly lower volumetric shrinkage while the bulk fill resin composites had significantly higher volumetric shrinkage than the conventional resin composites. It is concluded that each category of resin composite has different mechanical properties and polymerization behavior.

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### Conflict of Interest

The authors of this manuscript certify that they have no proprietary, financial, or other personal interest of any nature or kind in any product, service, and/or company that is presented in this article.

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