Effects of Thermal Cycling on Surface Texture of Restorative Composite Materials

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The purpose of this study was to evaluate the effect of thermal cycling on the surface texture of restorative materials. Disk-shaped specimens made of seven resin composites (Beautifil: BF; Esthet-X: EX; Filtek Supreme: FS; Inten-S: IS; Point 4: PT; Solare: SR; and Venus: VS) were finished with 1-μm alumina suspension, and then thermocycled between 4 and 60 ºC in distilled water for 20,000 or 50,000 cycles with a dwell time of 60 seconds. Staining susceptibility and mean surface roughness, Ra, were examined, and surface texture was observed by scanning electron microscopy. Dye penetration test showed that the surfaces of all resin composites were more stained after thermal cycling. Mean Ra of all resin composites, except PT, significantly increased after 50,000 thermal cycles. Dislodgement of filler particles was observed for all resin composites after thermal cycling, except FS. It was concluded that thermal cycling significantly affected the surface texture of the seven examined resin composites.

Keywords: Thermal cycling, Resin composite, Surface texture

INTRODUCTION

The last decade has witnessed significant improvements in the physical and mechanical properties, esthetics, and durability of resin composite materials for direct restorations1,2). Progress in bonding technology between tooth structures and filling materials has also enhanced the esthetics and prolonged the longevity of restored teeth3-5). With a focus on both the esthetics and longevity of restorations, various polishing methods to obtain optimum finishing surface — even highly polished surfaces — have been developed6-8).

However, predictably and inevitably, once a restorative material is exposed to the oral environment for a long period of time, it will be subjected to degradation in the esthetical aspect: staining, plaque accumulation, gingival irritation, recurrent decay, discoloration, and loss of gloss1,2,6-10). In addition, factors which may affect the surface roughness of resin composite restorative materials have also been investigated. In particular, toothbrush abrasion11-13), tooth-bleaching agents14-15), and prophylaxis materials16-19) have been found to increase surface roughness. As for the effect of pH on surface roughness, it was reported to be material-dependent13,20).

In the normal oral functioning of eating and drinking, restorative materials are also exposed to thermal stress. However, the effects of thermal cycling on the surface texture of resin composite restorative materials are yet to be comprehensively clarified. It was thus hypothesized by the authors of this paper that thermal cycling might influence the surface texture of resin composite restorative materials.

The purpose of this in vitro study, therefore, was to assess the effect of thermocycling on the surface roughness of some esthetic restorative materials. In this connection, staining susceptibility was also evaluated and surface topography of the materials was observed using scanning electron microscopy.

MATERIALS AND METHODS

Materials used
Table 1 lists the esthetic restorative composite materials used in this study — one giomer (Beautifil: BF; Esthet-X: EX; Filtek Supreme: FS; Inten-S: IS; Point 4: PT; Solare: SR; and Venus: VS) were finished with 1-μm alumina suspension, and then thermocycled between 4 and 60 ºC in distilled water for 20,000 or 50,000 cycles with a dwell time of 60 seconds. Staining susceptibility and mean surface roughness, Ra, were examined, and surface texture was observed by scanning electron microscopy. Dye penetration test showed that the surfaces of all resin composites were more stained after thermal cycling. Mean Ra of all resin composites, except PT, significantly increased after 50,000 thermal cycles. Dislodgement of filler particles was observed for all resin composites after thermal cycling, except FS. It was concluded that thermal cycling significantly affected the surface texture of the seven examined resin composites.

Keywords: Thermal cycling, Resin composite, Surface texture

Specimen preparation
Each composite material was packed into a Teflon mold with a hole of 8.0 mm diameter and 2.5 mm height between two microscope slides. The composite material was then photopolymerized with a light curing unit (Jetlite 1000, J Morita USA, Irvine, CA, USA) by irradiating 40 seconds for both sides. After which, specimens were stored in distilled water at...
for 24 hours, polished with 600-, 1,000-, 1,500-, and 2,000-grit silicon carbide papers, and finally finished with 1-μm alumina suspension into 2.00.05 mm thickness. Twenty-seven specimens were fabricated for each of the composite materials tested, and divided randomly into three groups (n = 9). One group was not thermocycled, and the remaining two groups were subjected to thermocycling for 20,000 or 50,000 cycles, respectively, between 4°C and 60°C water baths with one-minute dwell time at each temperature. On the overall, a total of 189 specimens for 21 groups, i.e., three groups for seven composite materials, were prepared.

Dye penetration test
Dye penetration test was carried out to determine the effect of thermocycling on the staining susceptibility of each composite material. From each group, one out of nine specimens was used for the dye penetration test. Each specimen was immersed separately in 5% methylene blue solution at 37°C for 24 hours. Then, specimens were rinsed with distilled water for 10 seconds and air-dried. The discoloration of each specimen was compared among the composite materials and among the thermocycling regimes by macroscopic observation.

Surface roughness measurement
Mean surface roughness (Ra) was measured for each thermocycling regime. Measurements for seven of the remaining eight specimens were performed using a profilometer (Surfcom 130A, Tokyo Seimitsu Co. Ltd., Tokyo, Japan). All readings were based upon a sampling length of 0.8 mm with measurement range of 40 μm and cut-off value at 0.8 mm. Three successive measurements in different directions were recorded for the seven specimens of each thermocycling regime.

The mean Ra of each specimen was calculated and statistically analyzed by one-way ANOVA for each group. Following which, the mean Ra of each group was determined for all the resin composites tested, and then statistically analyzed by two-way ANOVA with thermocycling number and composite material as independent factors. Differences among thermocycling regimes and among composite materials were analyzed by Bonferroni-Dunn test at

Table 1 Materials used in this study

<table>
<thead>
<tr>
<th>Material</th>
<th>Filler composition</th>
<th>Filler size</th>
<th>Filler content</th>
<th>Matrix resin composition</th>
<th>Manufacturer</th>
<th>Lot No.</th>
<th>Code</th>
</tr>
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<tbody>
<tr>
<td>Beautfil</td>
<td>F-B-Al-Si glass</td>
<td>0.01 - 5 μm</td>
<td>81.5</td>
<td>Bis-GMA/TEGDMA</td>
<td>Shofu, Kyoto, Japan</td>
<td>120381</td>
<td>BF</td>
</tr>
<tr>
<td>S-PRG</td>
<td></td>
<td></td>
<td>66.3</td>
<td></td>
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<tr>
<td>Esthet-X</td>
<td>Ba-Al-F-B-Si glass</td>
<td>0.02 - 2.5 μm</td>
<td>77</td>
<td>Bis-GMA adduct</td>
<td>Dentsply Caulk, Milford, DE, USA</td>
<td>0405034</td>
<td>EX</td>
</tr>
<tr>
<td>SiO₂</td>
<td>10 - 20 nm</td>
<td></td>
<td>60</td>
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<tr>
<td>Filtek Supreme</td>
<td>Zirconia-silica</td>
<td>5 - 75 nm (filler)</td>
<td>78.5</td>
<td>Bis-GMA/UDMA</td>
<td>3M ESPE, St. Paul, MN, USA</td>
<td>4AHJ</td>
<td>FS</td>
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<td>0.6 - 1.4 μm (cluster)</td>
<td></td>
<td></td>
<td>59.5</td>
<td>TEGDMA/Bis-EMA</td>
<td></td>
<td></td>
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<td>Inten-S</td>
<td>Barium glass/Silica/</td>
<td>0.2 - 0.7 μm</td>
<td>81.9</td>
<td>Bis-GMA/UDMA/</td>
<td>Ivoclar Vivadent, Schaan, Lichtenstein</td>
<td>F34199</td>
<td>IS</td>
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<tr>
<td>Titanium oxide</td>
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<td></td>
<td>51</td>
<td>Bis-EMA</td>
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<tr>
<td>Point 4</td>
<td>SiO₂</td>
<td>0.04 - 0.4 μm</td>
<td>77</td>
<td>Bis-GMA/TEGDMA</td>
<td>Kerr, Orange, CA, USA</td>
<td>308929</td>
<td>PT</td>
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<tr>
<td>Ba-Al-Si glass</td>
<td></td>
<td></td>
<td>59</td>
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<tr>
<td>Solare*</td>
<td>SiO₂</td>
<td>73</td>
<td>UDMA</td>
<td>GC, Tokyo, Japan</td>
<td>0401281</td>
<td>SR</td>
<td></td>
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<tr>
<td>Venus</td>
<td>Ba-Al-B-F-Si glass</td>
<td>0.7 μm / &lt; 2 μm</td>
<td>78</td>
<td>Bis-GMA/TEGDMA</td>
<td>Heraeus Kulzer, Hanau, Germany</td>
<td>010107</td>
<td>VS</td>
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<tr>
<td>SiO₂</td>
<td>0.01 - 0.07 μm</td>
<td></td>
<td>61</td>
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</tbody>
</table>

S-PRG: Pre-reacted glass ionomer (surface reaction type)
Bis-GMA: Bisphenol-A-glycidyl methacrylate
TEGDMA: Triethylene glycol dimethacrylate
Bis-GMA adduct: Adduct of 2,2-bis[(4-{2-hydroxy-3-methacryloyloxy propoxy) phenyl] propane with hexamethylene diisocyanate
Bis-EMA: Ethoxylated bisphenol-A-glycidyl methacrylate
UDMA: Urethane dimethacrylate

* Details of Solare composite material have never been published.
95% confidence level.

**SEM observation**
For the last remaining one of the nine specimens for each group, it was used for surface texture examination by using a scanning electron microscope (JSM-5510LV, JEOL, Tokyo, Japan).

**RESULTS**

**Dye penetration test**
Figure 1 shows the results of dye penetration test. Macroscopic observation revealed that all the resin composites became more stained as the number of thermocycles increased. The most stained surfaces were observed for Beautifil and Filtek Supreme, and the least stained surface was found for Inten-S.

**Surface roughness**
Figure 2 shows the mean Ra and standard deviation of each group, and the results of statistical analysis. Two-way ANOVA indicated significant differences among the resin composites (p<0.0001) and thermocycling regimes (p<0.0001) for the mean Ra of each group. In addition, significant interaction between resin composite and thermocycling (p<0.0001) indicated that some resin composites were more affected by thermocycling.

The mean Ra of each resin composite increased after thermocycling, except Point 4. For Beautifil, Inten-S, and Solare, the mean Ra significantly increased (p<0.0001) as the number of thermocycles increased. The mean Ra of Esthet-X and Venus after 50,000 cycles were significantly higher (p<0.0001) than those of their non-thermocycled counterparts. For Filtek Supreme, the mean Ra after 50,000 cycles was significantly higher (p<0.0001) than both non-thermocycled and 20,000-thermocycles groups. With Point 4, there were no significant differences among the mean Ra of the three thermocycling regimes.
Fig. 3 SEM micrographs of each resin composite before and after thermocycling.
In terms of comparison among resin composites at each thermocycling regime, Point 4 consistently showed the smoothest surface, whereas Inten-S revealed the roughest.

**SEM observation**

Figure 3 shows the SEM micrographs of seven resin composites after thermocycling. The SEM micrographs of the composites without thermocycling were also presented as a control.

All the materials showed a smooth surface before thermocycling, although some surface pitting was observed except with Filtek Supreme. For Filtek Supreme, filler particles were exposed after thermocycling, but pits as a result of dislodgement of filler particles were not observed.

Surface texture of Inten-S was similar to that of Solare. With Inten-S, filler particles in the resin matrix were dislodged after 20,000 cycles, and some of filler particles in the prepolymerized particles were also dislodged after 50,000 cycles. With Beautiful, large-sized pits as a result of loss of S-PRG filler particles were found after thermocycling. Furthermore, separation between filler particles and resin matrix, as well as pitting which might be due to the dissolution of filler particles, were also observed. As for the three microhybrid composite materials (Esthet-X, Point 4, and Venus), loss of filler particles was observed. Indeed, more filler particles were dislodged as the number of thermocycles increased.

**DISCUSSION**

The hypothesis in this study was that thermal cycling would influence the texture of resin composite restorative materials. Based on the results obtained with all the materials tested in this study, this hypothesis could be largely accepted. By dye penetration test, it was revealed that the surfaces became more stained after thermocycling. In terms of mean surface roughness, this aspect of each material significantly increased after 50,000 thermocycles, except Point 4. Dislodgement of filler particles was also observed after thermocycling, except with Filtek Supreme.

In previous studies that investigated composite restorative materials finished and polished with various finishing and polishing systems, the mean surface roughness ranged from 0.084 to 1.216 \( \mu \text{m} \). As the specimens used in this study were finally finished with 1-\( \mu \)m alumina suspension, surface roughness of non-thermocycled specimens ranged from 0.039 to 0.074 \( \mu \)m. These values were similar to those reported by Heintze and Forjanic, which were obtained by polishing with SiC abrasive paper and followed by 0.05-\( \mu \)m alumina suspension. However, the alumina particle size of 0.05\( \mu \)m was smaller than that used during dental treatments. Thus, the use of 1-\( \mu \)m alumina suspension in this study might have been more appropriate to provide comparable initial roughness as well as for intrinsic roughness evaluation of each material.

Some studies have evaluated the biological effects of different restorative materials on periodontal tissue. Willershausen et al. suggested that gingival irritation caused by resin-based materials might be due to technical errors in clinical practice or might arise from chemical properties of the materials. Paolantonio et al., on the other hand, reported that the negative effects of composite resin restorations on the quantity and quality of subgingival plaque might be caused by surface deterioration of composite restorative materials. According to Bollen et al., surface roughness exceeding a threshold value of 0.2\( \mu \)m would result in a simultaneous increase in plaque accumulation, thereby increasing the risk of caries and periodontal inflammation. In the present study, Ra values acquired for all the composite materials were less than 0.2\( \mu \)m, even after 50,000 thermal cycles. However, it should be put into perspective that many factors such as polishing method, toothbrush abrasion, tooth-bleaching agents, prophylaxis materials, pH derived from dietary acid, or the combinations thereof, may lead to a rougher surface exceeding the threshold value.

No direct relationship was found between mean surface roughness and staining susceptibility. Although the most stained surfaces were visually observed for Beautiful and Filtek Supreme after 50,000 thermal cycles (Fig. 1), their mean surface roughness values were not necessarily the highest. Conversely, the least stained surface was found for Inten-S. However, it consistently showed the roughest surface throughout the study. It should be highlighted that these results agreed with a previous study.

Discoloration of composite materials may be caused by both intrinsic and extrinsic factors. Intrinsic factors involve the discoloration of the resin material itself, such as alteration of resin matrix or degradation in the bonding interface between matrix and fillers. In other words, the type of resin matrix used in composite materials plays an important role in stain susceptibility. It has been reported that UDMA showed lower water sorption than Bis-GMA, and hence resulted in higher stain resistance. By this account, Inten-S and Solare, which included UDMA in the resin matrix, revealed the least stained surface. Filtek Supreme also contained UDMA in its resin matrix. However, due to the presence of TEGDMA which
increases water uptake in resin matrix\(^1^{,20}\). Filtek Supreme exhibited the most stained surface.

Extrinsic factors for discoloration include staining by adsorption or absorption of colorants as a result of contamination from exogenous sources\(^1\). Although a few materials have been identified as colorants \(\Box\) namely, methylene blue solution, coffee, tea, and wine, staining susceptibility was found to vary according to resin composite composition, polishing method, and colorant type\(^1^{,20}\). Therefore, the use of a different dye material in this study might have brought about different results.

During thermal cycling, the resin matrices of resin composite specimens would have absorbed some water\(^20\). It was also reported that water absorption would be accompanied by hygroscopic expansion in resin matrix and filler phase, thereby enhancing the weakening of matrix-filler interface. Then, due to difference in thermal expansion coefficient or thermal conductivity coefficient between resin matrix and filler particles, thermal cycling also caused repetitive shrinkage and expansion, resulting in a difference in thermal volumetric changes between resin matrix and filler particles. Furthermore, it must be mentioned that hydrolytic degradation of bonding between resin matrix and filler particles\(^{20}\) also occurred during thermal cycling. All these factors thus led to the dislodgement of filler particles. This phenomenon was observed for all materials tested in this study, except Filtek Supreme. With Filtek Supreme, agglomerated particles \(\Box\) which are called nanoclusters\(^{10}\) \(\Box\) were observed, and they seemed to resist plucking-out from matrix. Degradation of Filtek Supreme surface might have thus been caused by dislodgement of nanofillers from nanoclusters.

As for the mean surface roughness of each resin composite after thermal cycling, it could be attributed to several factors: filler loading, filler particle size, and adhesion between filler particles and resin matrix.

Beautifil was categorized as a hybrid material of pre-reacted glass ionomer (PRG) and resin composite. PRG involves the acid-base reaction of fluoroaluminosilicate glass fillers with polyacrylic acid. The resulting glass ionomer is freeze-dried, ball-milled, and silanized to form PRG fillers. These fillers are then incorporated in a resin matrix\(^{20,31}\). However, glass ionomer-like behaviors in terms of water absorption and hygroscopic expansion were long recognized as its biggest drawbacks\(^{31}\). As a result, Beautifil showed the most stained surface, and plucking-out of PRG fillers then led to rather high surface roughness values.

Inten-S was a heterogeneous hybrid composite material, whereby prepolymerized glass-filled particles were incorporated to reduce polymerization contraction\(^1^{,20}\). Solare was a microfine hybrid composite material which included prepolymerized organic filler particles. In the present study, surface texture of Inten-S was found to be similar to that of Solare. However, mean surface roughness of Inten-S was significantly higher than that of Solare after 50,000 thermal cycles. This phenomenon might have been caused by the dislodgement of micro glass filler particles from prepolymerized filler particles for Inten-S. As for Esthet-X, Point 4, and Venus, they were categorized as microhybrid composites. With increase in the number of thermocycles, more filler particles were dislodged and more pits were observed for these three materials. By SEM observation, the mean filler particle size of Esthet-X seemed to be the largest among the three materials, whereas that of Point 4 seemed to be the smallest. This finding was in good agreement with the mean surface roughness results.

With a view to evaluating the effects of temperature changes during eating and drinking on surface texture of resin composites for restorations, this in vitro study sought to simulate the thermal stress environment by means of thermal cycling. Indeed, for the purpose of evaluating the longevity of materials under conditions that more closely approximate clinical situations, further investigations are needed. While awaiting these future studies to be undertaken, another factor that significantly affects the longevity of restorative materials is bonding between filler particles and resin matrix \(\Box\) which definitely needs to be improved.

**CONCLUSIONS**

Within the limitations of this in vitro study, the following conclusions were drawn:

1. Thermal cycling was one important factor which affected the surface texture of resin composites for restorations.
2. By means of macroscopic observation after dye penetration test, it was revealed that all the resin composites became more stained as the number of thermocycles increased.
3. Mean surface roughness of each resin composite significantly increased after 50,000 thermocycles, except Point 4.
4. SEM observation showed the loss of filler particles after thermocycling for all resin composites, except Filtek Supreme.

**REFERENCES**

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