Effect of water storage and thermocycling on light transmission properties, translucency and refractive index of nanofilled flowable composites

Waleed ALMASABI1, Antonin TICHY1,2, Ahmed ABDOU1,3, Keiichi HOSAKA1, Masatoshi NAKAJIMA1 and Junji TAGAMI1

1 Cariology and Operative Dentistry, Graduate School of Medical and Dental Sciences, Tokyo Medical and Dental University, Tokyo, Japan
2 Institute of Dental Medicine, First Faculty of Medicine of the Charles University and General University Hospital in Prague, Prague, Czech Republic
3 Biomaterials Department, Faculty of Dentistry, Modern University for Technology and Information, Cairo, Egypt

Corresponding author, Masatoshi NAKAJIMA; E-mail: nakajima.ope@tmd.ac.jp

The effect of 1-week water storage and subsequent 10,000 thermal cycles on light transmission properties (straight-line transmission (G0), diffusion (DF), the amount of transmitted light (AV)), translucency parameter (TP) and refractive index (RI) of four nanofilled flowable composites was examined. The composites included Filtek Supreme Ultra Flowable Restorative (FSU), Estelite Flow Quick (EFQ), Estelite Universal Flow, (EUF), and Clearfil Majesty ES Flow (ESF), all of A3 shade. For composites with lower filler load (FSU, EFQ), water storage increased G0, AV and TP, but subsequent thermocycling decreased them. An opposite tendency was found for DF. Materials with higher filler load (EUF, ESF) were not significantly affected by aging conditions. RI of EFQ and EUF containing bisphenol A polyethoxy methacrylate (Bis-MPEP) increased significantly after thermocycling. Additionally, morphological changes were observed using scanning electron microscopy which revealed cracks within nanocluster fillers and dislocation of particles in FSU and EFQ after thermocycling.

Keywords: Nanocomposites, Fillers, Optical properties, Water storage, Thermal cycling

INTRODUCTION

With the development of adhesive systems, resin composite materials have been increasingly used for direct anterior and posterior restorations because of their improved physical and mechanical properties and esthetic appearance11. Resin composites are composed of an organic polymeric matrix, inorganic and/or organic fillers and a coupling agent that links the aforementioned components together. It is well known that mechanical properties, e.g., strength, hardness or fracture toughness, polishability and gloss are strongly influenced by filler particle characteristics, i.e., their shape, size, distribution and load2-5).

Owing to advances in nanotechnology, nanofilled flowable composites have recently been introduced to the dental market. The nanosized fillers are categorized into two types: (1) individually dispersed particles of size ranging between 5–100 nm, and (2) loosely bound agglomerates of nanoparticles (so-called nanoclusters) whose size exceeds 100 nm. The incorporation of nanosized filler particles led to superior esthetic properties and excellent wear resistance in addition to easy handling6-8). Compared to hybrid composites, nanofilled composites exhibited higher initial gloss and lower abrasive wear, however, they did not provide any advantage over hybrid composites in terms of strength and hardness9. It was also demonstrated that nanocluster fillers contributed to a greater water uptake and increased the susceptibility to fracture compared to conventional fillers10,11). Furthermore, several studies reported a loss of nanoparticles from the composite surface after long-term water storage or thermocycling1,12). Similarly to conventional fillers, silanization of the nanofillers is necessary prior to their incorporation into the resin matrix, so it has been speculated that non-optimized silane coupling treatment might lead to the detachment of isolated nanofiller and/or the dismantlement of nanoclusters4,13).

Fillers are also known to affect the optical properties of resin composites. When light is incident on a resin composite, it is partly reflected from the surface, while the passing light can be absorbed or scattered at the surface of the filler particles and diffused in multiple directions. Consequently, light can be transmitted through the composite either in the straight line or diffused. Light transmission is also affected by the mismatch of filler and matrix refractive indices14), so the light scattering characteristics of composites would be also dependent on the filler/matrix interface. If the link between them was degraded, water could penetrate into the gap between the filler particle and matrix, thus altering the scattering of incident light. Therefore, the degradation at the interface of the matrix and isolated particles and/or inside nanoclusters might affect the light transmission properties of nanofilled composites, especially light diffusion. Consequently, it might also affect their translucency which was previously shown to be significantly correlated with light diffusion15). This is clinically relevant, because the alteration in translucency of resin composites over time would deteriorate the esthetic appearance of resin-composite restorations. However, to our knowledge, no study has examined the effects of water storage or thermocycling.
on light transmission properties and translucency of nanofilled composites.

Immersion of resin composites in water leads to water sorption and solubility as a result of hydrolytic degradation of ester linkages in the polymer chain of resin matrix\(^{10}\). The uptake of water into resin matrix and elution of unreacted monomers might affect light transmission properties and the refractive index of resin composites as well. Therefore, the purpose of this study was to investigate the effects of water sorption and additional thermocycling stress on light transmission properties (straight-line light transmission (G0), light diffusion (DF) and the amount of transmitted light (AV)), translucency parameter (TP) and refractive index (RI) of nanofilled flowable resin composites. In addition, the microstructure of the composites was observed using scanning electron microscopy (SEM). The null hypotheses tested were that neither water sorption nor thermocycling would affect the light transmission properties (G0, DF, AV), TP and RI of the tested nanofilled flowable composites.

**MATERIALS AND METHODS**

Four nanofilled flowable resin composites of A3 shade were used in this study; Filtek Supreme Ultra Flowable Restorative (FSU; 3M, St. Paul, MN, USA), Estelite Flow Quick (EFQ; Tokuyama Dental, Tokyo, Japan), Estelite Universal Flow, (EUF; Tokuyama Dental), and Clearfil Majesty ES Flow (ESF; Kuraray Noritake Dental, Tokyo, Japan). Their composition is listed in Table 1.

**Specimen preparation**

Plastic molds (6 mm diameter, 0.5 mm thickness) were placed on glass slides covered with celluloid strips. The flowable resin composites were applied to the molds, covered with another celluloid strip and glass slide, and polymerized for 60 s from both sides using a quartz-tungsten halogen light-curing unit (Optilux 501, 530 mW/cm\(^2\); Demetron, Danbury, CT, USA). The discs (n=7) were polished using 2000-grit silicon carbide paper and stored in dry conditions and darkness for 24 h, followed by 1-week storage in distilled water at 37°C, and 10,000 thermal cycles (TC) between 5°C and 55°C (dwell time 30 s, transfer time 2 s). The measurements of G0, DF, AV, TP and RI were performed immediately after polishing, after 24 h, 1-week water storage, 5,000 TC and 10,000 TC.

**Measurement of light transmission properties**

The measurements were performed using a goniophotometer (GP-200, Murakami Color Research Laboratory, Tokyo, Japan) under standardized conditions (sensitivity: 950; volume: 522) without filter. The incidence angle was set to 0° and two-dimensional distribution graphs of transmitted light intensities were obtained in the range of −90° to +90°. The peak gain at 0° angle was used to calculate G0, while DF was calculated using following formula: 

\[
DF(\%) = \frac{(B_{50} + B_{20})}{2} \times 100, \\
\]

where \( B \) is the light intensity at a certain angle. The total area of the distribution graph corresponding to AV was calculated using the Image J software (version 1.47 for Windows, National Institutes of Health, Bethesda, MD, USA).

**Measurement of translucency**

The translucency of resin composites is an important factor for the visual appearance of restorations and masking of background color. It can be assessed using several methods, e.g. translucency parameter (TP) or contrast ratio\(^{10}\). These parameters are known to be

---

**Table 1  Composition of tested materials**

| Materials (shade, abbreviation) | Manufacturer (batch number) | Resin matrix | Filler composition | Filler content |
|---------------------------------|-----------------------------|--------------|--------------------|---------------|
| Filtek Supreme Ultra Flowable Restorative (A3, FSU) | 3M, St. Paul, MN, USA (N897475) | Bis-GMA, TEGDMA, Procrylat resin | silica (20 nm, 75 nm), zirconia (5–10 nm), zirconia/silica clusters (0.6–10 µm), ytterbium fluoride (0.1–5 µm) | 65 wt% (46 vol%) |
| Estelite Flow Quick (A3, EFQ) | Tokuyama Dental, Tokyo, Japan (J2782) | Bis-MPEPP, TEGDMA, UDMA | silica-zirconia supra-nano spherical filler (0.07 µm, 0.4 µm) | 71 wt% (53 vol%) |
| Estelite Universal Flow (A3, EUF) | Tokuyama Dental (202) | Bis-GMA, Bis-MPEPP, TEGDMA, UDMA | spherical silica-zirconia filler (200 nm), prepolymerized filler (200 nm) | 71 wt% (57 vol%) |
| Clearfil Majesty ES Flow (A3, ESF) | Kuraray Noritake Dental, Tokyo, Japan (9T0203) | TEGDMA, hydrophobic aromatic dimethacrylate | silanated barium glass filler, silanated silica nanocluster filler (0.18–3.5 µm) | 75 wt% (59 vol%) |

Bis-GMA: bisphenol A-glycidyl methacrylate, TEGDMA: triethylene glycol dimethacrylate, Procrylat: (2,2-bis[4-(3-methacryloxypropoxy)phenyl]propane), Bis-MPEPP: bisphenol A polyethoxy methacrylate, UDMA: urethane dimethacrylate
strongly correlated\textsuperscript{18,19}, so only TP was measured in this study, because it corresponds directly to common visual assessments of translucency and descriptions of the masking ability of materials\textsuperscript{17}.

The composite discs were placed on black and white backings, and their color was measured using a non-contact reflection spectrophotometer (CrystalEye, Olympus, Tokyo, Japan) equipped with 7 LEDs as an illumination source. The spectral data of a selected area (2.0×2.0 mm) in the center of each specimen were analyzed to determine the CIELAB co-ordinates \( L^*, a^*, b^* \). TP was obtained by calculating the color difference of the specimen placed on black (\( B \)) and white (\( W \)) backings according to the formula: \( TP = [(L_B^*-L_W^*)+(a_B^*-a_W^*)+(b_B^*-b_W^*)]^{1/2} \).

The use of a coupling medium such as glycerin was proposed to mimic wet conditions in the oral cavity more closely\textsuperscript{18}, but to our knowledge, this is still not a standard procedure. In this study, no coupling medium was placed between the backings and the composite specimens to avoid their contamination which could affect the outcomes of other tests. Although nominal values of TP with and without glycerin differ, they were shown to be strongly correlated\textsuperscript{18}, so relative differences between various storage conditions examined in this study should not be altered.

**Measures of refractive index**

Swept-source optical coherence tomography (SS-OCT) has been previously used to measure the refractive index of hard dental tissues\textsuperscript{20} and dental materials\textsuperscript{21}. In this study, the measurements were performed using IVS-2000 (Santec, Komaki, Japan) with a laser light source sweeping a bandwidth of 100 nm (central wavelength 1,330 nm) at a rate of 30 kHz. The axial resolution in air was 12 µm, while the lateral resolution was 17 µm. The laser beam scans the specimens in X and Z dimensions and the backscattered light detected by the system is digitized in time scale. Subsequently, the data are analyzed in the Fourier domain to produce depth-resolved scans (A-scans) and cross-sectional scans (B-scans). The Image J software (version 1.47 for Windows, National Institutes of Health) was used to analyze the B-scans and to determine the optical path length (OPL). Since OPL is the product of \( RI \) and the actual specimen thickness (\( t \)), \( RI \) was calculated using the equation\textsuperscript{20}: \( RI = \frac{OPL}{t} \).

**Statistical analysis**

Kolmogorov-Smirnov and Shapiro-Wilk tests were used to examine the distribution normality of the G0, DF, AV, TP and RI data. Since the tests indicated that all distributions were normal, a repeated measures ANOVA with Bonferroni correction was used for each tested parameter to compare between different flowable composites and aging conditions at \( a=0.05 \). Greenhouse–Geisser correction was used for the lack of sphericity in tested parameters. The analyses were performed using the IBM SPSS software (Version 23, IBM, Armonk, NY, USA).

**Scanning electron microscopy**

Four additional discs were prepared for each composite as described in section 2.1. Half of the discs (\( n=2 \)) were stored in dry conditions and darkness for 24 h, while the other half (\( n=2 \)) were stored in distilled water at 37°C for 1 week and subjected to 10,000 TC between 5°C and 55°C. After the respective storage conditions, the specimens were polished to high gloss using diamond pastes (DP-Paste P, Struers, Copenhagen, Denmark) with particle size decreasing from 6 µm to 0.25 µm, desiccated, and sputter-coated with gold. The morphology of the composite surfaces was observed using a scanning electron microscope (JSM-IT100, JEOL, Tokyo, Japan) at magnification 25,000×.

**RESULTS**

**Light transmission properties**

The values of G0 are presented in Table 2, DF in Table 3, and AV in Table 4. Repeated measures ANOVAs revealed that the effects of different materials and aging conditions were strongly significant (\( p<0.001 \)) for all three parameters. Their interaction was significant for G0 (\( p<0.001 \)), but not for DF (\( p=0.620 \)) and AV (\( p=0.186 \)).

EFQ exhibited the highest G0 of the tested materials, followed by FSU. Compared to the immediate and 24-h values, the G0 of EFQ and FSU increased significantly after the 1-week water storage (\( p<0.001 \)). However, the values after 10,000 TC were significantly lower than the equation:

\[
G0 = \frac{I1 - I2}{I1} \times 100
\]

where \( I1 \) is the light transmission of the composite in dry condition, \( I2 \) is the light transmission of the composite in wet condition.

Table 2  Straight-line light transmission (G0) results

| Material | Immediate | 24 h   | 1 week | 5,000 TC | 10,000 TC |
|----------|-----------|--------|--------|----------|-----------|
| FSU      | 27.8±4.8\textsuperscript{Aa} | 32.1±5.9\textsuperscript{Aa} | 41.6±8.4\textsuperscript{Ab} | 29.2±5.7\textsuperscript{Aa} | 28.9±7.0\textsuperscript{Ab} |
| EFQ      | 56.0±11.0\textsuperscript{Bb} | 63.4±14.4\textsuperscript{Bb} | 74.8±14.6\textsuperscript{Bc} | 75.0±14.3\textsuperscript{Bc} | 57.2±17.5\textsuperscript{Bb} |
| EUF      | 8.7±1.6\textsuperscript{Ca}  | 8.5±1.0\textsuperscript{Ca}  | 8.6±1.7\textsuperscript{Ca}  | 7.8±1.1\textsuperscript{Ca}  | 8.4±1.6\textsuperscript{Ca}  |
| ESF      | 14.0±3.4\textsuperscript{Da} | 15.0±4.5\textsuperscript{Da} | 16.8±4.9\textsuperscript{Da} | 16.0±6.0\textsuperscript{Da} | 17.1±7.3\textsuperscript{Da} |

Different superscript uppercase letters indicate significant differences (\( p<0.05 \)) in columns, lowercase letters in rows. 24 h: 24-h dry storage, 1 week: 1-week water storage, TC: thermal cycles, FSU: Filtek Supreme Ultra Flowable Restorative, EFQ: Estelite Flow Quick, EUF: Estelite Universal Flow, ESF: Clearfil Majesty ES Flow
Table 3  Light diffusion (DF) results

|       | Immediate | 24 h       | 1 week      | 5,000 TC    | 10,000 TC   |
|-------|-----------|------------|-------------|-------------|-------------|
| FSU   | 17.1±2.2
|       |           | 14.6±1.6  | 15.3±1.6  | 15.3±1.2  | 20.9±7.0
|       |           |           |             |             |             |
| EFQ   | 17.4±1.6  | 16.2±2.2  | 16.0±2.2  | 18.5±3.7  | 18.3±4.0  |
|       |           |           |             |             |             |
| EUF   | 34.8±1.3  | 33.8±1.7  | 35.6±1.2  | 37.3±5.3  | 36.6±4.2  |
|       |           |           |             |             |             |
| ESF   | 31.5±1.4  | 30.0±2.0  | 31.7±1.5  | 34.0±6.1  | 34.7±5.5  |

For the interpretation of significant differences and the explanation of abbreviations, please refer to Table 2.

Table 4  The amount of transmitted light (AV) results

|       | Immediate | 24 h       | 1 week      | 5,000 TC    | 10,000 TC   |
|-------|-----------|------------|-------------|-------------|-------------|
| FSU   | 16,108±1,778 | 16,467±4,003 | 19,571±2,205 | 16,234±2,183 | 16,907±1,316 |
|       |           |           |             |             |             |
| EFQ   | 17,797±1,308 | 17,682±1,493 | 2,148+1,812 | 19,281±2,413 | 19,568±1,961 |
|       |           |           |             |             |             |
| EUF   | 11,693±2,075 | 10,882±903 | 11,823±2,395 | 10,559±2,172 | 11,562±2,498 |
|       |           |           |             |             |             |
| ESF   | 10,810±1,007 | 9,842±1,299 | 12,146±1,855 | 10,642±3,310 | 12,372±2,924 |

For the interpretation of significant differences and the explanation of abbreviations, please refer to Table 2.

Table 5  Translucency parameter (TP) results

|       | Immediate | 24 h       | 1 week      | 5,000 TC    | 10,000 TC   |
|-------|-----------|------------|-------------|-------------|-------------|
| FSU   | 27.0±1.9  | 29.0±2.8  | 29.9±2.7  | 27.4±1.8  | 25.8±1.8  |
|       |           |           |             |             |             |
| EFQ   | 30.7±2.6  | 31.3±1.6  | 31.5±1.0  | 32.4±2.0  | 31.1±2.3  |
|       |           |           |             |             |             |
| EUF   | 26.7±4.2  | 26.2±2.5  | 26.1±2.1  | 24.6±0.9  | 26.8±1.2  |
|       |           |           |             |             |             |
| ESF   | 26.2±0.9  | 25.4±2.8  | 26.2±3.1  | 26.4±1.5  | 24.8±3.5  |

For the interpretation of significant differences and the explanation of abbreviations, please refer to Table 2.

Table 6  Refractive index (RI) results

|       | Immediate | 24 h       | 1 week      | 5,000 TC    | 10,000 TC   |
|-------|-----------|------------|-------------|-------------|-------------|
| FSU   | 1.39±0.02 | 1.39±0.04 | 1.40±0.04 | 1.38±0.05 | 1.42±0.02 |
|       |           |           |             |             |             |
| EFQ   | 1.35±0.03 | 1.34±0.04 | 1.37±0.05 | 1.36±0.05 | 1.40±0.01 |
|       |           |           |             |             |             |
| EUF   | 1.36±0.04 | 1.37±0.02 | 1.39±0.04 | 1.37±0.03 | 1.42±0.01 |
|       |           |           |             |             |             |
| ESF   | 1.38±0.02 | 1.40±0.04 | 1.39±0.04 | 1.38±0.05 | 1.40±0.04 |

For the interpretation of significant differences and the explanation of abbreviations, please refer to Table 2.

than after 1 week ($p<0.017$ for EFQ and $p=0.001$ for FSU) and they were not significantly different from the immediate values. The G0 values of EUF and ESF were not significantly influenced by aging conditions ($p>0.959$ and $p>0.717$, respectively).

The DF of EUF and ESF were not significantly different from each other, but significantly higher than the values of FSU and EFQ. Aging conditions had no significant effect on the DF of EFQ, EUF and ESF ($p>0.05$). The DF of FSU decreased significantly after 24 h ($p=0.002$) and then gradually increased during the 1-week water storage and thermocycling, in which there was a significant difference between values after 5,000 and 10,000 TC ($p<0.05$).

The AV of FSU and EFQ were statistically similar and significantly higher than the values of EUF and ESF. EUF and ESF were not significantly influenced by aging conditions ($p>0.703$ and $p>0.105$, respectively). On the other hand; the 1-week water storage significantly increased the AV of FSU and EFQ ($p<0.001$) and their values decreased after thermocycling, but significantly only for FSU after 10,000 TC ($p=0.045$).
Translucency parameter
The repeated measures ANOVA revealed that TP (Table 5) was significantly affected by different materials ($p<0.001$) but not by aging conditions ($p=0.156$). The interaction between materials and aging conditions was significant ($p=0.02$). EFQ exhibited the highest TP of the tested materials. Aging conditions did not significantly influence the TP of EFQ, EUF and ESF. In contrast, FSU exhibited an insignificant increase in TP after 1 week of water storage ($p=0.077$) followed by a significant decrease in TP after 10,000 TC compared to the 1-week values ($p=0.009$).

Refractive index
The repeated measures ANOVA disclosed that RI (Table 6) was significantly influenced by aging conditions ($p<0.001$), whereas the effect of materials was not significant ($p=0.156$) as well as their interaction ($p=0.482$). There were no significant differences in RI between the tested materials. The RI of FSU and ESF were not influenced by aging conditions ($p>0.089$ and $p>0.172$, respectively). For EFQ and EUF, RI increased significantly after 10,000 TC compared with the immediate values ($p=0.004$ and $p=0.019$, respectively).

SEM analysis
Representative SEM images of the tested materials after 24-h dry storage and 10,000 TC at magnification 25,000× are presented in Fig. 1. Nanoclusters of size surpassing 1 µm were observed in FSU (Figs. 1A, B) and cracks were observed in the nanoclusters and at the filler/matrix interface after thermocycling (Fig. 1B). The observation of EFQ revealed two types of regular round-shaped particles of approximately 0.3 µm and <0.1 µm size, respectively (Figs. 1C, D). After thermocycling, some of the particles were dislocated (Fig. 1D). Round-shaped particles of sub-micrometer size were observed in EUF (Figs. 1E, F) and EFQ (Figs. 1G, H), and no apparent difference was observed at their surfaces after thermocycling.

**DISCUSSION**

The optical properties of resin composite are determined by many factors, such as resin matrix composition, filler load, size, composition and content, pigments and other additives. Additionally, they are also affected by differences in refractive indices of resin matrix and fillers whose mismatch may decrease the translucency of resin composites. In this study, the effects of storage in water for 1 week and subsequent thermocycling on light transmission properties (G0, DF, AV), TP and RI of four commercial nanofilled flowable composites were examined. The 1-week water storage significantly increased the G0 and AV values of FSU and EFQ, but a significant decrease after 10,000 TC followed except for the AV of EFQ. Thermocycling also significantly increased the RI of EFQ and EUF compared to their immediate values. Therefore, the null hypotheses were partially rejected.

When immersed in water, resin composites absorb water and the sorption extent is influenced by the composition and volume of resin matrix. Generally, the lower the filler load in resin composites, the higher the water sorption. In this study, the 1-week water storage significantly increased the G0 and AV values of FSU and EFQ, but a significant decrease after 10,000 TC followed except for the AV of EFQ. Thermocycling also significantly increased the RI of EFQ and EUF compared to their immediate values. Therefore, the null hypotheses were partially rejected.
of light passing through the composites. In contrast, light transmission properties of EUF and ESF with higher filler load (57 and 59 vol%, respectively) were not significantly affected, presumably due to a lower extent of water sorption.

Following the 1-week water storage, specimens were subjected to thermocycling which induces mechanical stress by cyclic thermal expansion and shrinkage. As a consequence of different thermal expansion coefficients of fillers and resin matrix, microcracks could develop within the matrix and/or nanocluster fillers. Moreover, long-term exposure to higher temperature could enhance the hydrolytic degradation of silane coating on filler surface and thus cause debonding at the filler/matrix interface. Depending on the number of thermal cycles, the detachment of fillers from the matrix at the surface could increase the surface roughness of the composites which might lead to increased light scattering/absorption at the surface and hence decreased amount of transmitted light (AV). In this study, 10,000 TC did not significantly change AV compared with the values after 1-week water storage except for FSU whose AV decreased significantly after 5,000 TC and remained similar after 10,000 TC. Although surface roughness was not measured in this study, these results suggest that it was not significantly affected by thermocycling.

This is in agreement with a previous report that 10,000 TC did not significantly affect the surface roughness of nanohybrid and microhybrid resin composites. Nevertheless, some significant differences in G0, DF, and TP were observed with FSU and EFQ after 10,000 TC, indicating increased light scattering/absorption inside of the composites rather than at the surface.

In the case of FSU, 5,000 TC significantly decreased G0 and AV compared to the values after 1-week water storage which might be due to the light scattering/absorption at the surface. On the other hand, a significant increase in DF was observed between 5,000 and 10,000 TC although AV did not change, suggesting intrinsic changes in the material. In addition, the TP of FSU gradually decreased with the increasing number of TC, and its inverse relationship with DF corroborates the results of a previous study which demonstrated their negative correlation. FSU contains nanoclusters, which are loosely bound aggregates of zirconia and silica nanoparticles, and the SEM observation of the re-polished thermocycled specimens revealed microcracks within the nanoclusters as well as their debonding from the resin matrix (Fig. 1B). The detachment of isolated nanofiller from the matrix and/or the dismantlement of nanoclusters in the FSU and ESF were not significantly affected by thermocycling.

The light transmission parameters and TP of ESF and EUF were not significantly affected by thermocycling. Furthermore, no obvious morphological alterations were revealed using SEM (Figs. 1E–H). ESF contains nanocluster fillers composed of silica nanoparticles treated with a highly hydrophobic silane coupling agent, so we presume that the silanization contributed to the high resistance of nanoclusters in ESF to the hydrolytic stress during water storage and mechanical stress induced by thermocycling. EUF contains silica-zirconia spherical nanoparticles and prepolymerized fillers composed of an organic polymerizable resin with a high load of the silica-zirconia nanoparticles. Owing to the industrial polymerization and high filler load, we suppose that prepolymerized fillers are very resistant to external stresses and that they contributed to the stability of EUF.

The RI of all tested materials slightly increased after the immersion in water for 1 week and a further increase was observed after thermocycling. As a result, the RI of EFQ and EUF after 10,000 TC were significantly different from the immediate values. EFQ and EUF contain Bis-MPEPP (bisphenol A polyethoxy methacrylate), a monomer whose side-chains are lacking the presence of hydroxyl groups, which might lead to less internal cross-linking compared to Bis-GMA (bisphenol A-glycidyl methacrylate). As a consequence, Bis-MPEPP-based composites would be less stable, thus increasing the entrapment of water within the polymer network. Lee et al. also demonstrated that Bis-MPEPP-based composites leached more substances than Bis-GMA and Bis-GMA/UDMA-based composites. We speculate that the elution of monomers from the Bis-MPEPP-based composites EFQ and EUF and their replacement with absorbed water could cause the increase in their RI after water storage and thermocycling.

CONCLUSION

Within the limitations of this study, it was concluded that water sorption decreased light scattering and increased the amount of light transmitted through nanofilled flowable resin composites with lower filler load. Thermocycling had an opposite effect on light transmission properties due to the degradation of nanocluster fillers and the filler/matrix interface. Silanated and prepolymerized fillers were more resistant.
to degradation and the change of light transmission properties and translucency. The refractive index of resin composites increased after water storage and thermocycling, presumably due to the elution of unreacted monomers.

ACKNOWLEDGMENTS
This work was supported by Ministry of Education, Culture, Sports, Science and Technology of Japan (grant numbers 18K09571 and 19K10106).

CONFICT OF INTEREST
None.

REFERENCES
1) Ilie N, Hickel R. Resin composite restorative materials. Aust Dent J 2011; 56: 59-66.
2) Jun SK, Kim DA, Goo HJ, Lee HH. Investigation of the correlation between the different mechanical properties of resin composites. Dent Mater J 2013; 32: 48-57.
3) Elbishari H, Silikas N, Satterthwaite JD. Effect of filler properties and translucency. The refractive index of resin composites affected by filler size? Int J Dent 2020; 2020: 1-6.
4) Lohbauer U, Belli R, Ferracane JL. Factors involved in mechanical fatigue degradation of dental resin composites. J Dent Res 2013; 92: 584-591.
5) Ornaghi BP, Meier MM, Lohbauer U, Braga RR. Fracture toughness and cyclic fatigue resistance of resin composites with different filler size distributions. Dent Mater 2014; 30: 742-751.
6) de Oliveira GU, Mondelli RFL, Charantola Rodrigues M, Franco EB, Ishikiriama SK, Wang L. Impact of filler size and distribution on roughness, gloss and color of composite resin after simulated toothbrushing. J Appl Oral Sci 2012; 20: 510-516.
7) Hayashi S, Homma S, Takanashi T, Hirano T, Yoshinari M, Yajima Y. Wear properties of esthetic dental materials against translucent zirconia. Dent Mater J 2019; 38: 256.
8) Lee IB, Chang J, Ferracane J. Slumping resistance and viscoelasticity prior to setting of dental composites. Dent Mater 2008; 24: 1586-1593.
9) Alzaikat H, Burrow MF, Maghaireh GA, Taha NA. Nanofilled resin composite properties and clinical performance: A review. Oper Dent 2018; 43: e173-e190.
10) Curtis AR, Shortall AC, Marquis PM, Palin WM. Water uptake and strength characteristics of a nanofilled resin-based composite. J Dent 2008; 36: 186-193.
11) Curtis AR, Palin WM, Fleming GP, Shortall ACC, Marquis PM. The mechanical properties of nanofilled resin-based composites: The impact of dry and wet cyclic pre-loading on bi-axial flexure strength. Dent Mater 2009; 25: 188-197.
12) Minami H, Hori S, Kurashige H, Murarahara S, Muruguchi K, Minesaki Y, et al. Effects of thermal cycling on surface texture of restorative composite materials. Dent Mater J 2007; 26: 316-322.
13) Nagano D, Nakajima M, Takahashi M, Ikeda M, Hosaka K, Sato K, et al. Effect of water aging of adherend composite on repair bond strength of nanofilled composites. J Adhes Dent 2018; 20: 425-433.
14) Shortall AC, Palin WM, Burtscher P. Refractive index mismatch and monomer reactivity influence composite curing depth. J Dent Res 2008; 87: 84-88.
15) Arimoto A, Nakajima M, Hosaka K, Nishimura K, Ikeda M, Foxton RM, et al. Translucency, opalescence and light transmission characteristics of light-cured resin composites. Dent Mater 2010; 26: 1090-1097.
16) Ferracane JL. Hygroscopic and hydrolytic effects in dental polymer networks. Dent Mater 2006; 22: 211-222.
17) Johnston WM. Review of translucency determinations and applications to dental materials. J Esthet Restor Dent 2014; 26: 217-223.
18) Nogueira AD, Della bona A. The effect of a coupling medium on color and translucency of CAD-CAM ceramics. J Dent 2013; 41 Suppl 3: e18-e23.
19) Barizon KTL, Bergeron C, Vargas MA, Qian F, Cobb DS, Gratton DG, et al. Ceramic materials for porcelain veneers. Part I: Correlation between translucency parameters and contrast ratio. J Prosthodont Dent 2013; 110: 397-401.
20) Hariri I, Sadr A, Shimada Y, Tagami J, Sumi Y. Effects of structural orientation of enamel and dentine on light attenuation and local refractive index: An optical coherence tomography study. J Dent 2012; 40: 387-396.
21) Kazuko Y, Yu K, Mikihiro K, Mai Y, Hiromi O, Takashi M, et al. Refractive index measurement of dental materials by swept-source optical coherence tomography. Jpn J Conserv Dent 2018; 61: 368-377.
22) Arikawa H, Kanie T, Fujii K, Takahashi H, Ban S. Effect of filler properties in composite resins on light transmission characteristics and color. Dent Mater J 2007; 26: 38-44.
23) Lee YK. Influence of filler on the difference between the transmitted and reflected colors of experimental resin composites. Dent Mater 2008; 24: 1243-1247.
24) Biradar B, Biradar S, Ma A. Evaluation of the effect of water on three different light cured composite restorative materials stored in water: An in vitro study. Int J Dent 2012; 2012: 1-5.
25) Dos Santos PH, Catelan A, Guedes APA, Suzuki TYU, Godas AGDL, Briso ALF, et al. Effect of thermocycling on roughness of nanofill, microfill and microhybrid composites. Acta Odontol Scand 2015; 73: 176-181.
26) Rinastiti M, Özcan M, Siswomihardjo W, Busscher HJ. Effects of surface conditioning on repair bond strengths of non-aged and aged microhybrid, nanohybrid, and nanofilled composite resins. Clin Oral Investig 2011; 15: 625-633.
27) Pala K, Tekce N, Tuncer S, Serim ME, Demirci M. Evaluation of the surface hardness, roughness, gloss and color of composites after different finishing/polishing treatments and thermocycling using a multitechnique approach. Dent Mater J 2016; 35: 278-289.
28) Lee SY, Huang HM, Lin CY, Shih YH. Leached components from dental composites in oral simulating fluids and the resultant composite strengths. J Oral Rehabil 1998; 25: 575-588.