Effects of rotating fatigue on the mechanical properties of microhybrid and nanofiller-containing composites

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The objective of this study was to assess and compare the flexural strength and Vickers hardness of five nanofiller-containing composites (Filtek Supreme XT, Gradia Forte, Luna-Wing, GNH400N, GCUC) against five microhybrid composites (Meta Color Prime Art, Solidex, Estenia C&B, Ceramage, Clearfil Majesty) before and after rotating fatigue test (RFT). For each resin composite, 16 rectangular beam specimens (2 mm×2 mm×25 mm) were prepared and half of which were subjected to 1×10⁴ cycles in RFT. Flexural strength was determined using a three-point bending test. Vickers hardness measurements were carried out on specimens which failed after the three-point bending test. When under the influence of rotating fatigue, the flexural strength of all composites was affected by multiple factors. In contrast, rotating fatigue had no significant influence on the Vickers hardness of both microhybrid and nanofiller-containing composites.

Keywords: Nanofiller-containing composite, Rotating fatigue testing, Flexural strength, Vickers hardness

INTRODUCTION

Dental resin composites are favored among the dentists and widely used in esthetic restorative treatments. Similarly, they have attracted considerable interest among researchers. The effects and impacts of their physical properties, polymerization shrinkage behavior, handling properties, durability, and wear resistance on clinical performance have been extensively investigated. While considerable research attention was focused on clarifying the influences of methacrylate resin matrix, filler type, and filler loading on material properties — and hence the associated clinical outcome, the properties of individual inorganic filler, when isolated from the organic matrix, also deserve thorough research and investigation.

New technological advancements in filler technology have caused the dental market to be inundated with a plethora of filler types, filler morphologies, and particle size distributions, posing a great challenge to the characterization of fillers in modern dental composites. A recent development is the application of nanotechnology in direct dental restorative materials. The growing number of dental composites containing nanofiller particles reflects the growing interest in nanostructured dental composite restorative materials.

Resin-based dental composites are increasingly being used in direct posterior restorations. However, evidence from studies had shown that their long-term clinical performance in high-load-bearing posterior restorations was inferior compared to amalgam. The physical properties and mechanical behavior of dental composite restorative materials are generally determined under static or quasi-static loading. Under clinical circumstances, however, composite restorations in posterior teeth are subjected to cyclic loading from mastication, grinding, and shivering. For an average person, cyclic stresses are repeated more than 3×10⁵ times per year. Repetitive loading due to mastication can cause fatigue and lead to subcritical crack propagation in tooth tissue and/or restorations, resulting in failure at stress levels significantly lower than the ultimate bond strength.

When comparing existing composite restorative materials or designing new ones, it is important to take cyclic fatigue into consideration. The rotating cantilever beam test has been used to evaluate the fatigue resistance of resin-based composites. It uses the staircase (up-and-down) method to determine fatigue limits. After initial stress was set at about 50% of the fracture strength calculated for a group of specimens, a specimen would be rotated for a predetermined number of cycles (10,000 or 1,000,000) or until failure occurred. If failure occurred before reaching the predetermined number of cycles, the test would be repeated with a new specimen at the initial stress level minus a predetermined force increment. If the specimen survived, the test would be repeated at the initial stress level plus a predetermined force increment.
force increment. These iterations which characterized the up-and-down staircase pattern thus compared the test groups at different fatigue stress levels.

Little is known about the fatigue resistance of resin-based composites at the same stress level using the rotating fatigue test (RFT). The objective of this study was to assess and compare the flexural strength and Vickers hardness of five nanofiller-containing composites against five microhybrid composites before and after RFT.

**MATERIALS AND METHODS**

**Specimen preparation**

Five microhybrid composites (Meta Color Prime art, Solidex, Estenia C&B, Ceramage, Clearfil Majesty) and five nanofiller-containing composites (Filtek Supreme XT, Gradia Forte, Luna-Wing, GNH400N, GCUC) were evaluated in this study. They were listed in Table 1 according to their commercial product names and manufacturers, alongside details such as composition and filler content according to manufacturer data and release documents.

According to ISO specification 4049\(^\text{18}\), 16 rectangular beam specimens (2 mm×2 mm×25 mm) of each resin composite were prepared using a stainless steel split mold, and half of which were subjected to RFT. After unpolymerized resin was placed in the mold, specimen was pressed between two glass plates and light-cured from two sides for 90 s each using a light curing unit (Twin Cure, Shofu, Kyoto, Japan). Before each use, light intensity was measured to verify that curing power remained constant at 800 mW/cm\(^2\).

After hardening, resin flash at the edges was removed. The edges were then beveled with 1,200-grit silicon carbide grinding paper (Buehler, Tokyo,

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**Table 1  Microhybrid and nanofiller-containing composites used in this study**

<table>
<thead>
<tr>
<th>Product/Code/Batch/Manufacturer</th>
<th>Filler content (wt%)</th>
<th>Main composition</th>
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<tbody>
<tr>
<td><strong>Microhybrid composites</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Meta Color Prime Art/MCPA/TL6/Sun Medical (Shiga, Japan)</td>
<td>73</td>
<td>UDMA, TEGDMA, aromatic amine, silica, others</td>
</tr>
<tr>
<td>Solidex/SD/070928/Shofu (Kyoto, Japan)</td>
<td>78</td>
<td>UDMA, organic filler, silica power</td>
</tr>
<tr>
<td>Estenia C&amp;B/ECB/0022BA/Kuraray Medical (Tokyo, Japan)</td>
<td>87.9</td>
<td>Bis-GMA, UDMA, UTMA, ultrafine alumina particles (average particle size of 20 nm), fine aluminosilicate glass particles (average particle size: 1.5 μm)</td>
</tr>
<tr>
<td>Ceramage/CMG/050994/Shofu (Kyoto, Japan)</td>
<td>73</td>
<td>UDMA, zirconium silicate</td>
</tr>
<tr>
<td>Clearfil Majesty/CM/00221A/Kuraray Medical (Tokyo, Japan)</td>
<td>81</td>
<td>Bis-GMA, TEGDMA, hydrophobic aromatic dimethacrylate, silanated barium glass filler, prepolymerized organic filler</td>
</tr>
</tbody>
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<table>
<thead>
<tr>
<th>Nanofiller-containing composites</th>
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<tbody>
<tr>
<td>Filtek Supreme XT/FS/20090708/3M ESPE (St. Paul, MN, USA)</td>
<td>78.5</td>
<td>Bis-GMA, Bis-EMA, UDMA, TEGDMA, zirconia and silica particles (clusters of 0.6–1.4 μm, individual particle size of 5–20 nm)</td>
</tr>
<tr>
<td>Gradia Forte/GF/0909081/GC (Tokyo, Japan)</td>
<td>75</td>
<td>UDMA, silica power, prepolymerized filler</td>
</tr>
<tr>
<td>Luna-Wing/LW/01070922/Yamakin (Osaka, Japan)</td>
<td>68</td>
<td>Methacrylate monomer, inorganic filler</td>
</tr>
<tr>
<td>(Trial production) GNH400N/GC (Tokyo, Japan)</td>
<td>85</td>
<td>N/A</td>
</tr>
<tr>
<td>(Trial production) GCUC/GC (Tokyo, Japan)</td>
<td>83</td>
<td>N/A</td>
</tr>
</tbody>
</table>

UDMA: urethane dimethacrylate, UTMA: urethane tetramethacrylate, Bis-EMA: bisphenol-A-polyethylene glycol diether dimethacrylate, Bis-GMA: 2,2'-bis[4(2-hydroxy-3-methacryloyloxy-propoxy)-phenyl]propane, TEGDMA: triethyleneglycol dimethacrylate, N/A: Not available
Japan). Using a digital caliper (ABS Digimatic Caliper, Mitutoyo, Kanagawa, Japan), specimen thickness was measured to ensure that only specimens of 2.0±0.05 mm thickness were accepted. Each specimen was further inspected on a light box to ensure the absence of porosity. All specimens were stored in 37°C distilled water for 24 h prior to use.

Three-point bending test
To determine the flexural strength of composite specimens before RFT, a three-point bending test was conducted according to ISO 4049, using a universal testing machine (Servopulser EHF-FD1, Shimadzu, Kyoto, Japan) at a crosshead speed of 0.5 mm/min until fracture occurred. Flexural strength was calculated according to the formula below:

$$\sigma = \frac{3F_{\text{max}}l}{2bh^2}$$ \hspace{1cm} (1)

where $F_{\text{max}}$ is the load at fracture (N), $l$ is the length between supports (15 mm), and $b$ and $h$ are the width (2 mm) and height (2 mm) of each specimen.

Vickers hardness measurement
Vickers hardness measurements were carried out on fractured specimens acquired from the three-point bending test. Using the digital caliper, site of hardness measurement was measured to be 8 mm from the free end of the rectangular beam specimen. A 10-kg load was applied on specimen surface for 15 s using a diamond pyramid indenter (AVK-A, Akashi, Hyogo, Japan). Dimensions of each indentation produced were measured as the lengths of the diagonals of the indentation mark at a microscopic level, and each measurement was repeated three times. Vickers hardness was calculated using the formula below:

$$HV = \frac{1.854P}{d^2}$$ \hspace{1cm} (2)

where $P$ is the force applied (N) and $d$ is the arithmetic mean of the two indentation diagonals.

Finite element analysis
According to Mirmohammadi et al.\textsuperscript{12} in their investigation on the influence of rotating fatigue, finite element (FE) analysis indicated that the generated tensile stress reached peak value when one corner of the rectangular beam specimen came into the top position. Therefore, in this study, FE analysis was also used to calculate the maximum tensile stress to be applied to the composite specimens in RFT.

Figure 1 shows a 2-dimensional (2D) FE model of the crown of a premolar with a mesio-occlusal-distal (MOD) composite restoration. This model, which had been used by Shi et al.\textsuperscript{19}, was created from the CT images of a real tooth. Using a FE mesh generation procedure (PLANE42, ANSYS Version 12.0, Ansys Inc., Houston, PA, USA), the 2D model created contained 1,344 4-node elements, 1,415 nodes, and a thickness set at 1 mm. Material properties were assumed to be isotropic and linearly elastic. For the composite restoration, enamel, dentin, and the pulp, their Young’s modulus values were 10, 84, 20, and 0.002 GPa respectively; the Poisson’s ratios used were 0.24, 0.30, 0.31, and 0.45 respectively\textsuperscript{20}. Perfect bonding was assumed between the tooth and the composite restoration. To identify the most highly stressed regions within the composite restoration, the restoration was created with a much finer mesh than the other areas of the 2D model, with an average element edge length of 0.1 mm. The model was fully constrained at the lower boundary, which was close to the enamel-dentin junction. Occlusal loading of 100 N was simulated in FE analysis program (ANSYS Version 12.0) using a rigid ball of 4.5 mm diameter.

Figure 2 shows the FE analysis results. Maximum tensile stresses were generated on the occlusal surface of MOD composite restoration. Maximum tensile stress value was about 25.7 MPa. To calculate the constant load ($F$) to be applied to the composite beam specimen with one of its corners in the top position, the following equation\textsuperscript{12} was used:

$$F = \frac{Sb^3}{6\sqrt{2\times l}}$$ \hspace{1cm} (3)

where $S$ (MPa) is the maximum tensile stress, $l$ is the distance between the loading point and the rotating grip (10.0 mm), and $b$ is the width of the beam (2.0 mm).

Based on Eq. (3), composite beam specimens in RFT would be loaded with constant load $F$ at 242 g.

Rotating fatigue test
In the mouth, forces on a tooth or restoration may be
applied in a buccolingual, occlusoapical, or mesiodistal direction, or even a combination of these force vectors. In RFT, specimen was additionally rotated by 180 degrees to complete a 360-degree circle to simulate these multivectorial force patterns encountered in the mouth.

Figure 3 shows the cantilever beam test setup for RFT. One end of the rectangular beam specimen was clamped in a rotating grip. The other free end was loaded through a ball bearing by the constant force of a coil spring. Based on FE analysis and calculation using Eq. (3), a constant load of 242 g was applied.

During RFT, the specimen was subjected to repeated tension-compression stress cycling as it rotated from zero degrees (maximum compression) to 180 degrees (maximum tension), then back to zero degrees again. All specimens were rotated at 1.2 Hz for 10,000 cycles. If failure occurred before reaching 10,000 cycles, the fractured specimen was replaced by a new one.

Statistical analysis
Means and standard deviations were calculated for all 10 composite groups. One-way analysis of variance (ANOVA), Tukey’s post hoc test, and independent t-test were used to analyze their flexural strength and Vickers hardness data by means of a computer software (SPSS Version 15.0, SPSS Inc., Chicago, IL, USA). P-values less than 0.05 were considered to be statistically significant.
RESULTS

Mechanical properties

Table 2 summarizes the mean flexural strength and Vickers hardness values of each resin composite before and after RFT.

1. Flexural strength

Before RFT, the highest mean flexural strengths were found in GF (192.6±11.8 MPa), GCUC (175.8±12.7 MPa), and ECB (175.3±18.5 MPa). The next intermediate group consisted of GNH400N, CMG, and CM. Lowest mean flexural strengths were found in LW, FS, SD, and MCPA.

After RFT, the highest mean flexural strengths were found in GF (177.2±9.9 MPa), ECB (167.4±18.1 MPa), GCUC (166.9±12.2 MPa), and GNH400N (165.5±17.2 MPa). Two FS specimens and one MCPA specimen failed before reaching 2,000 cycles.

2. Vickers hardness

Before RFT, the highest mean Vickers hardness was found in EBC (185.2±20.2 Hv). There were no significant differences in Vickers hardness before and after RFT for each composite.

DISCUSSION

Rotating fatigue test

The rotating fatigue test reproduces the repetitive, alternating, and multivectorial force pattern encountered in the mouth. It exerts multivectorial stress on its test specimens in a sequence of tension and compression in each cycle12). Given the contrast between the multivectorial loading pattern performed in rotating fatigue test and the monotonic loading performed in three-point bending test, several researchers have reported on the discernible gaps between rotating fatigue resistance and static three-point flexural strength. Scherrer et al.21) reported that the rotating fatigue resistance of composites was 40% to 60% of the monotonic flexural strength determined by three-point bending test. Other studies comparing cyclic to static loading also showed similar ratios of rotating fatigue resistance to static strength in the range of 50% to 70%12,22).

In the conventional rotating test which uses the up-and-down staircase analysis method, mean fatigue resistance is obtained using the stress level at which 50% of the specimens failed. In the monotonic three-point bending test, flexural strength is obtained when 100% of the specimens failed. Besides, there typically exists considerable variation and scatter in the flexural strength for many specimens of a brittle composite.
Fig. 5 SEM image (1,000×) of the filler particles of MCPA.

Fig. 4 SEM images (10,000×) of the morphology and size of filler particles separated from the resin matrix. Most filler particles are irregular-shaped. Spherical particles are found in SD, CMG, and FS. Nanoclusters (indicated by arrows) are found in FS, GNH400N, and GCUC.

material. For these reasons, a constant load was used in this study in place of increasing or decreasing stress levels characteristically used in the conventional staircase method.

It could be argued that in the case of up-and-down staircase method, the raw data collected is non-normally distributed and requires data manipulation by appropriate statistical analysis methods. However, data manipulation and analysis only makes interpretation of the experimental data more challenging with its complicated interpretation of the results. In essence, the authors concluded that there was no comparison between the incompatible data of rotating fatigue resistance and static flexural strength.

It remained to be examined whether the test data obtained in rotating fatigue tests were valid predictors of clinical performance. Nonetheless, this method clearly revealed differences in mechanical properties among the materials tested, which was the case in this study. To ensure that the rotating fatigue test faithfully
simulate the alternating and multivectorial intraoral loading pattern, an important question to answer was: what is the number of cycles which represents the clinical situation? No hard evidence exists concerning the number of chewing strokes annually. In published literature, the number of test cycles varied widely from \(10^2\) to \(10^6\) \cite{15-17,28}. A research on the relationship between stress and cycle numbers (S-N curves) of unidirectional glass/epoxy composites revealed that high low-cycle fatigue loads led to failure in less than \(10^4\) cycles \cite{18}. Therefore, \(10^4\) cycles was used in this study. Besides, a low-cycle fatigue of 0.25 Hz frequency resulted in a lower survival rate than high-cycle fatigue of 5–10 Hz frequency \cite{18}. On a clinical basis, the upper limit of the chewing frequency is assumed to be two strokes per second\cite{24}. Putting these together, the rotation frequency used in this study was 1.2 Hz.

Although teeth are three-dimensional structures, a premolar with a MOD composite restoration within the buccolingual section is sufficient for the investigation of various important mechanical factors. Therefore, a 2D plane strain model was used for FE analysis in this study. The use of a 2D model also offers improved performance in terms of element shape and simulation quality. Biting force has been reported to be as high as 790 N\cite{25}, but this value is the so-called peak load. According to Anderson\cite{26,27}, mean masticatory force ranged between 70.6 and 146.1 N. For FE analysis in this study, the applied load was 100 N—an averaged value of the masticatory force as reported by Anderson\cite{26,27}. Maximum tensile stress, rather than maximum compressive stress, was calculated by FE analysis and applied in RFT. This was because dental composites displayed higher strengths in compression than in tension (on the order of a factor of 5–8)\cite{28}.

**Filler properties**

Generally, microfills are particles which are smaller than 1 μm and nanofills are particles which are smaller than 0.1 μm. In this study, SEM images revealed that most of the microhybrid composites contained particles which varied between 0.5 and 3 μm, while nanofiller-containing composites contained particles which varied between 0.05 and 0.5 μm. For example, MCPA—which is classified as a microhybrid composite\cite{29}—contained large fillers which exceeded 30 μm (Fig. 5). In the strict sense of particle size definition, MCPA could not be considered as a microhybrid composite. Similarly, the filler particles of LW and GF did not meet the given nano-size criterion; instead, they seemed to be more like microhybrid composites.

Spherical particles were found in SD, CMG, and FS. A spherical shape reportedly enhanced fracture strength since mechanical stresses tend to concentrate on the angles and protuberances of filler particles\cite{29}. In this study, however, SD and FS exhibited the lowest mean flexural strengths among the microhybrid and nanofiller-containing composites respectively. The authors concluded that the causative factors and reasons require further investigation.

Compared to ECB, GNH400N, and GCUC, SD and FS had a lower filler content—which could be one of the reasons accounting for their low flexural strengths. Predominantly, FS contained nanoclusters which were partially calcined and infiltrated with a silane coupling agent prior to incorporation into the resin matrix together with the dispersed individual nanoparticles\cite{31}. For GNH400N and GCUC, the nanoclusters seemed to be synthesized by a modified method which allowed higher filler loadings, which then resulted in higher flexural strengths than FS. FS also seemed to be more waxy during specimen preparation. This was probably due to the TEGDMA ingredient, which has been reported to reduce flexural strength.

**Effects on mechanical properties**

Results in Table 2 did not show a positive correlation between flexural strength and filler content. Both nanofiller-containing composite GF and microhybrid composite MCPA had a similar filler content (73–75 wt%), but they exhibited the highest and lowest flexural strengths respectively. For all the tested composites, their flexural strengths decreased after RFT, especially so for those with lower initial flexural strengths. Average rate of decrease was 27% for MCPA, 26% for SD, 13% for FS, and 12% for LW.

Among the microhybrid composites, ECB showed the highest flexural strength. ECB had a superior polymer matrix based on Bis-GMA and UDMA, which was further reinforced with aluminosilicate particles. Bis-GMA and UDMA are the most widely used base monomers in the polymeric matrices of dental composites, chiefly because they present superior mechanical properties. Bis-GMA is a bulky, high-molecular-weight monomer which has high strength and high modulus due to its aromatic ring structure. However, TEGDMA is a widely used diluent for Bis-GMA resins. Consequently, the handling and physical properties of dental composites are unavoidably and adversely influenced to some extent by the TEGDMA ingredient.

EBC also showed the highest mean Vickers hardness among all the tested composites. Previous studies have shown that the key to increasing the compressive strength of dental composites was to increase the filler content\cite{32,33}. The results of the present study further confirmed a strong correlation between hardness and filler content.

**CONCLUSIONS**

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

1. When under the influence of rotating fatigue, the flexural strength of both microhybrid and nanofiller-containing composites was affected by multiple factors and which could not be straightforwardly explained by filler morphology and particle size.
2. Irrespective of filler morphology and particle size,
the flexural strengths of all composites decreased after RFT, especially those with lower initial flexural strengths.

3. Rotating fatigue had no significant influence on the Vickers hardness of both microhybrid and nanofiller-containing composites.

REFERENCES


