Fatigue of Tooth-colored Restoratives in Aqueous Environment

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The purpose of this study was to investigate the interaction between mechanical and chemical fatigue in resin composites and dental ceramics, and the effects thereof on fatigue resistance of tooth-colored restoratives. To this end, the fatigue fracture resistance of restoratives under dry and aqueous conditions were analyzed by a dynamic fatigue crack propagation test using beam-shaped specimens with a precrack. Fatigue crack propagation characteristics were expressed by the correlation between fatigue crack growth rate (da/dN) and stress intensity factor range (ΔK). In addition, fatigue crack growth threshold (ΔKth) was calculated. Following the fatigue test, a fractographic examination was performed using scanning electron microscopy. Fatigue crack initiation was retarded in resin composites under aqueous condition, but dental ceramics were susceptible to slow crack growth after crack initiation. SEM images of the fatigue fracture surfaces reflected inorganic and organic filler particles of different sizes in composites and the bonding at crystal-glass interface in ceramics. It was concluded that water exerted different effects on the fatigue resistance of composites and ceramics.

Keywords: Fatigue, Dental ceramic, Resin composite

INTRODUCTION

In the oral environment, fatigue fracture of dental restoratives can be induced by relatively weak and repetitive loads, such as the force arising from ordinary chewing1-2. For resin composites, several mechanisms may participate in fatigue-induced damage of resin composite materials, which include matrix cracking, matrix deformation, void formation, multidirectional cracking, filler debonding, and filler failure3. For dental ceramics, experimental difficulties associated with their extreme brittleness have prevented their fatigue behavior to be sufficiently represented—but recent studies have proven that they are affected by both mechanical and chemical fatigue4-5. Fatigue fracture of resin composites and dental ceramics originates from an internal or external micro flaw, from which cracks propagate and eventually result in a complete catastrophic fracture6. However, inevitably, flaws that can trigger crack initiation are introduced during the fabrication of composites and ceramics as well as during the restoration procedure. Therefore, the characteristics of fatigue crack propagation, expressed in terms of fatigue crack growth rate (da/dN) and stress intensity factor range (ΔK), are appropriate for evaluating the fatigue resistance of dental restoratives. Likewise, it is appropriate for fatigue crack initiation to be expressed in terms of fatigue crack growth threshold (ΔKth).

Currently, dynamic fatigue crack propagation test for resin composites is conducted using a compact tension specimen in the form of a double cantilever beam11-13. However, a suitable fatigue crack propagation test for dental ceramics has not been established, largely due to the difficulty associated with fabricating a relatively large specimen with a complicated shape. Therefore, to date, only static fatigue test with indentation has been used to evaluate fatigue crack propagation in dental ceramics14-15. However, dynamic fatigue crack propagation tests are presumably more appropriate than static fatigue tests for evaluating the fatigue resistance of dental restoratives16, since fatigue resistance is overestimated under static loading17. Against this background, the present study set out to conduct the first dynamic fatigue crack propagation test for dental ceramics using beam-shaped specimens with a precrack, which were fabricated by a Compression Bridge Assembly (Fig. 1).

Aging in water or aqueous fluids is well known to decrease the fatigue resistance of polymer-based composites as a result of water absorption by the matrix18. However, inconsistencies in fatigue crack propagation behavior of composites under aqueous conditions have been reported19-21. In our previous investigation21, it was confirmed that contrary to common perception on fatigue fracture behavior of resin composites, fatigue crack propagation in resin composites was retarded under aqueous conditions but accelerated after water immersion. This indicated that the adverse effect of water on fatigue resistance of composites was dependent on experimental conditions. Moreover, fatigue cracks detoured...
around inorganic macrofillers but penetrated organic fillers, suggesting that inorganic macrofillers were instrumental to retarding fatigue cracks in resin composites.

In industrial ceramics, the deleterious effect of slow crack propagation can be attributed to a stress-enhanced chemical reaction, known as "stress corrosion", that occurs in the presence of water vapor at a crack tip\(^{16}\). However, the effects of water on dynamic crack propagation in dental ceramics have not been confirmed. Therefore, there is a need to investigate the interaction between mechanical and chemical fatigue in dental ceramics and clarify their respective roles in fatigue behavior in the oral environment, and thereby compare the results against resin composites.

The present study investigated the fatigue fracture mechanism of tooth-colored restoratives by conducting dynamic fatigue crack propagation test followed by fractographic examination. The null hypothesis in this study was that crack propagation in resin composites and dental ceramics was not affected by the presence of water.

**MATERIALS AND METHODS**

**Preparation of specimens**

Beam-shaped specimens measuring 3.0 \(\times\) 5.0 \(\times\) 10.0 mm were fabricated from resin composites and dental ceramics (Table 1). Specimens for heat-pressed and castable ceramics were fabricated according to the manufacturers’ instructions, and resin composite specimens were light-cured for 14 minutes in a custom-made metallic mold. All specimens were polished with 1500-grit SiC paper followed by 1-\(\mu\)m alumina particles on a water-irrigated grinding wheel (Ecomet III, Buehler Ltd., Lake Bluff, IL, USA) to eliminate superficial microflaws. The edges of all the beam specimens were ground.

Starter notches to introduce a precrack were created by applying a vertical load of 10 N for 30 seconds with a Knoop stylus, followed by an additional vertical load of 50 N for 10 seconds with a Vickers stylus. Two kinds of indentation were performed so as to obtain a precrack in a straight form. Three starter notches were then arranged at the center on the tension side of each beam specimen at 0.25-mm intervals (Fig. 1). Next, a precrack - which connected the starter notches and which propagated over the whole surface of the tension side with a maximum depth of 1 mm - was initiated by applying a compression load using a Compression Bridge Assembly (Fig. 1) according to JIS R 1607\(^{18}\). Extension of precrack was monitored under microscopic observation with the aid of transillumination, and the depth of precrack was confirmed under the microscope using dye penetration. All specimens were stored in a desiccator at room temperature or in water at 37 \(\degree\) for 48 hours before the fatigue test.

**Fatigue crack propagation test**

Fatigue crack propagation tests were performed in

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**Table 1** Resin composites and ceramics investigated in this study.

<table>
<thead>
<tr>
<th>Code</th>
<th>Product name (manufacturer)</th>
<th>Composition</th>
<th>Filler/crystal content (vol%)</th>
<th>Mean particle size of filler/crystal ((\mu)m)</th>
<th>Lot No.</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Resin composites</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>APX</td>
<td>Clearfil AP-X (Kuraray, Osaka, Japan)</td>
<td>Bis-GMA, TEGDMA, Barium glass, Silica</td>
<td>68-71</td>
<td>3</td>
<td>303</td>
</tr>
<tr>
<td>PAE</td>
<td>Palfique Estelite (Tokuyama, Tokyo, Japan)</td>
<td>Bis-GMA, TEGDMA, Barium zirconia filler, Organic filler</td>
<td>71</td>
<td>Inorganic filler: 0.2 Organic filler: 40</td>
<td>EU115</td>
</tr>
<tr>
<td><strong>Dental ceramics</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>EMP</td>
<td>IPS Empress (Ivoclar Vivadent, Schaen, Liechtenstein)</td>
<td>SiO(_2), Al(_2)O(_3), K(_2)O, Leucite crystals</td>
<td>24</td>
<td>1-5</td>
<td>1474</td>
</tr>
<tr>
<td>DCR</td>
<td>Dicor (Dentsply International, York, PA, USA)</td>
<td>K(_2)O-Mg(_2)O-MgO-SiO(_2), Fluoromica crystals</td>
<td>65</td>
<td>2</td>
<td>94708</td>
</tr>
</tbody>
</table>
general accordance with ASTM E647-93 using an
electro servohydraulic testing machine (Servopulser
EHF FD01, Shimadzu Co., Kyoto, Japan) in both dry
and aqueous environments. The test under dry con-
dition was conducted at room temperature (23 °C),
while that under aqueous condition was conducted in
a circulating water bath (Thermo Supplier EZL-80,
Taitek Co., Saitama, Japan) at 37 °C.

Sinusoidal cyclic loading was applied at a fre-
quency of 10 Hz with a mean stress ratio
(R=Kmin/Kmax) of 0.1. Length of the vertical crack
propagation was measured every 60 seconds by an
optical microscope (Traveling Microscope Type A,
Shimadzu Co.) at a magnification of ~40 with the
aid of trans-and reflecting illumination. The fatigue
test came to a completion when the crack propagated
through the whole vertical surface of the specimen.

Resistance to fatigue crack propagation was ex-
pressed in terms of fatigue crack growth rate (da/ 
dN) and crack tip stress intensity factor range (∆K),
as defined by the theory of linear elasticity and
shown in the following equation:

\[
\frac{da}{dN} = C(\Delta K)^m
\]

where \(a\) is the crack length, \(N\) the number of cyclic
loads, \(C\) and \(m\) the experimentally determined scaling
constants, and \(\Delta K\) the variation in stress intensity
factor in a cycle - which is as shown below:

\[
\Delta K = K_{\text{max}} - K_{\text{min}}
\]

As for fatigue fracture resistance, it was ex-
pressed in terms of fatigue crack growth threshold
(\(\Delta K_{\text{th}}\)) - which was the asymptotic value of \(\Delta K\) at
which \(da/dN\) approached zero. \(\Delta K_{\text{th}}\) was given as
the \(\Delta K\) value corresponding to a fatigue crack
growth rate of \(10^{-3}\) m/cycle.

Fractographic examination
After the fatigue crack propagation test, a vertical
compression load was applied by the servohydraulic
testing machine until a complete fracture occurred.
Fractured surfaces were then observed by scanning
electron microscopy (JSM9840A, JEOL Ltd., Tokyo,
Japan) at magnifications ranging from ~50 to ~1500 after sputter-coating with a gold-palladium
alloy.

RESULTS

Characteristics of fatigue crack growth in resin com-
posites and dental ceramics under dry and aqueous
conditions are shown in Fig. 2. The \(m\) values of
resin composites and dental ceramics were in the
ranges of 11.6-76.5 and 22.9-54.6 respectively. The

Fig. 2 Characteristics of fatigue crack propagation in resin composites and dental ceramics under dry (△) and aqueous (○) conditions.
Fatigue fracture and unstable fracture surfaces of resin composites and dental ceramics under aqueous condition. Left: area including the boundary (△) between fatigue and unstable fractures at low magnification; middle: brittle fracture surface at high magnification; right: fatigue fracture surface at high magnification.

Boundary between the fatigue and unstable fractures was evident; as well the fatigue fracture surface rougher than the unstable fracture surface in both composites and ceramics. Filler particles were clearly observed on the fatigue fracture surface in both resin composites. With APX, inorganic fillers with a diameter of 50 μm were exposed on the fatigue fracture surface. With PAE, fatigue fracture pathway was found through the prepolymerized organic fillers with diameter of 40 μm in some areas. Fluoromica crystals in DCR were detected all over the fatigue fracture surface, while leucite crystals in EMP were not.
\( K_{\alpha} \) values of resin composites and ceramics were calculated to be 0.51-1.08 and 0.63-1.07 respectively. Resin composites showed greater \( K_{\alpha} \) values under aqueous condition than under dry condition, whereas ceramics had smaller \( K_{\alpha} \) values under aqueous condition than under dry condition. In particular, the \( K_{\alpha} \) values in dry and aqueous conditions differed remarkably for APX and DCR.

Fractographic examination revealed no morphological differences in fracture surface between dry and aqueous environments. Representative fatigue fracture and unstable fracture surfaces of composites and ceramics under aqueous condition are shown in Fig. 3. Boundaries between fatigue and unstable fractures were evident, and fatigue fracture surfaces were rougher than unstable fracture surfaces in both composites and ceramics.

Filler particles were clearly observed at the fatigue fracture surfaces of both resin composites. In APX, inorganic fillers with a diameter of 5 \( \mu \)m were exposed on the fatigue fracture surface. In PAE, fatigue fracture pathway was found through the prepolymerized organic fillers with a diameter of 40 \( \mu \)m in some areas. In DCR, fluoromica crystals were detected all over the fatigue fracture surface, while leucite crystal sin EMP were not.

**DISCUSSION**

The fatigue resistance of dental ceramics is commonly expressed in terms of both crack initiation and propagation, since fatigue tests are generally conducted using plain specimens without precracks. This has been so because it is difficult to initiate a precrack in preparation for the fatigue test due to the extreme brittleness of dental ceramics. However, internal and external microflaws are inevitable in dental restoratives. As such, fatigue crack growth test using specimens with a precrack is considered more appropriate for evaluating clinical fatigue resistance. In the present study, crack initiation was successfully achieved by machining pyramid-shaped starter notches, and a precrack thereby obtained by propagating a crack from this region by applying compressive force using a Compression Bridge Assembly.

When cyclic compressive force was applied after storing resin composites in water or aqueous fluids for defined periods of time, the fatigue fracture resistance decreased compared with that under dry condition - findings which were consistent with the results of our previous study. Reasons for the inferior fatigue resistance of composites after storage in water were considered to be due to hydrolyses of the resin matrix and filler-matrix interface, arising from water absorption.

Contrary to common perception, the present study indicated that the fatigue fracture resistance of resin composites increased under aqueous condition - a finding consistent with our previous study too. A similar increase in fracture resistance of a carbon-coated graphite composite was also reported when specimens were completely immersed in water. The increased fracture resistance of composites under aqueous condition in the present study could be explained by the following phenomena due to water absorption by the resin matrix. Water absorption encourages plasticization of the resin matrix - resulting in decreased brittleness of the composites, as well as releases the internal tensile stress generated during polymerization shrinkage and the residual compressive stress at the crack tip. These phenomena then contributed to the retardation of crack propagation in composites under aqueous condition, either independently or in combination.

It is important to be aware of the adverse effect of water on fatigue resistance of composites - although it depends on the experimental conditions. While toughening of resin composites under aqueous condition was observed in the present study, long-term deterioration of composites due to water absorption has also been reported. In other words, interaction between these antagonistic mechanisms of retardation of crack propagation versus hydrolysis of the resin matrix can proceed simultaneously in resin composites, and the effect of such interaction on fatigue behavior after long-term water immersion requires further investigation.

In both composites, filler particles were clearly observed on the fatigue surface, suggesting that fatigue crack propagation pathway was present at the matrix-filler interface. In PAE, the crack pathway penetrated the organic fillers in some areas (Fig. 3), and their \( K_{\alpha} \) values were remarkably smaller than those of APX, regardless of the environmental condition. These observations indicated that organic fillers were less effective for retarding crack propagation in resin composites due to their inferior mechanical properties. Filler particle size in resin composites has been reported to be one of the critical factors for retarding fatigue crack growth. The results of the present study suggested that filler components - organic versus inorganic filler particles - also played a critical role against fatigue fracture resistance in composites.

Crystals in ceramics contribute to retardation of fatigue crack growth via the mechanisms of crack bridging, crack deflection, and welding - either independently or in combination. The leucite crystals in EMP could be considered to be useful for retarding cracks, despite their low content (24%), since EMP retained relatively high \( K_{\alpha} \) values even under aqueous condition. Further, fractographic examination revealed that leucite crystals could hardly be detected.
on the fatigue fracture surface, and that the glass matrix still adhered to the crystals. This cohesive fracture thus suggested firm crystal-matrix adhesion, which could render optimal strength properties against fatigue. In contrast, fluoromica crystals were observed all over the fatigue fracture surface of DCR, suggesting predominant adhesive failure between the crystals and the glass phase.

Slow crack growth kinetics are known to be strongly influenced by the amount and composition of the glass phase in a ceramic microstructure[5]. Stress corrosion preferentially occurs in silicate-based ceramics, resulting in bond rupture due to hydrolysis[6]. The binding of Si-O-Si at the tip of a crack splits into Si-OH and OH-Si under aqueous conditions with tensile stress. In the present study, bonding strength at the crystal-glass interface in DCR appeared to be less durable against fatigue fracture, particularly under the aqueous condition, despite a high crystal content (65%). This finding thus suggested that firm bonding between glass matrix and crystals was more critical for fracture resistance than the crystal content.

Taken together, the results of the present study demonstrated that water exerted different effects on fatigue fracture resistance in resin composites and dental ceramics. For resin composites, the bonding between the resin matrix and filler particles played a critical role against fatigue resistance, and likewise the bonding between the glass phase and crystals in dental ceramics.

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