The repair bond strength to resin matrix in cured resin composites after water aging

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The repair microshear bond strengths (µSBSs) to resin matrices in 4 different cured-composites after water storage (0, 60 s, 1 week, 1 month) were evaluated. Three different adhesive application methods to the cured-composites were performed; (1) none, (2) one-step self-etch adhesive application, and (3) one-step self-etch adhesive application with a silane coupling agent. Degree of conversion (DC) of the composite discs was determined using ATR/FT-IR with a time-based spectrum analysis. Initially, the amount of un-reacted resin monomers in the repaired cured-composite contributed to the bonding performance of newly-filled uncured-composite to resin matrix of the cured-composite. Adhesive application could not improve their repair µSBS. After 1-month of water-storage, the repair µSBS was dependent on material, which either reduced or did not and was not influenced by their amount of un-reacted resin monomers. When repairing aged composite resin, the appropriate adhesive application procedures were different among resin composites.

Keywords: Repair bond strength, Water aging, Resin matrix, Adhesive application, Degree of conversion

INTRODUCTION

Direct resin composite restorations are increasingly placed in dental practice because of their good esthetics and superior bonding performance to tooth substrates. In accordance with the concept of minimal intervention and maximum preservation of tooth structure, repair of resin composite restorations is recommended instead of the complete-removal and replacement in cases of secondary caries, marginal defects and discoloration or discoloration of the resin composite itself. The repair bonding performance between newly placed and aged polymerized resin composite restorations depends on three mechanisms: 1) chemical interaction with remaining un-reacted monomers in previously cured-composite; 2) micro-mechanical bonding by monomer infiltration into the cured-composite and chemical bonding to exposed fillers on the cured-composite surface. Initially, the quantity of remaining un-reacted resin monomers in cured-composites would be influenced by the polymerization rate. Therefore, resin composite materials with lower rates of polymerization would offer an advantage for obtaining higher immediate repair bonding performance whereas a higher polymerization rate would be disadvantageous. However, these un-reacted monomers leach from the cured-composite during aging in the oral environment and therefore their ability to positively influence repair bonding performance would be gradually lost. Moreover, the resin matrix in cured-resin composite absorbs water during aging. The presence of water absorbed in cured-composite could interfere with the infiltration of hydrophobic monomers from the newly-placed uncured composite into the cured-composite surface, leading to a reduction in the repair bonding performance. The application of a hydrophilic adhesive, such as a one-step self-etch adhesive may improve the repair bonding performance to water-aged cured-composites. There have been many studies about the pre-treatment effect of a silane coupling agent on repair bonding performance to water-aged cured-composites. However, there have been fewer investigations on repair bonding performance to the resin matrix of water-aged cured-composites. The objective of this study was therefore to evaluate repair bond strengths to the resin matrices of resin composites after water storage for 60 s, 1 week and 1 month.
month. The null hypotheses tested in this study were that 1) there is no difference in repair bond strength between resin composites, and that 2) water-storage does not affect the repair bond strength of resin composites with and without application of a one-step self-etch adhesive.

MATERIALS AND METHODS

Microshear bond strength (μSBS) test

The materials used in this study are listed in Table 1. Four resin composites, Filtek Supreme Ultra (FSU; Shade A2, 3M, St. Paul, MN, USA), Estelite Sigma Quick (ESQ; Shade A2, Tokuyama Dental, Tokyo, Japan), Beautifil II (BE2; Shade A2, Shofu, Kyoto, Japan), and Clearfil AP-X (APX; Shade A2, Kuraray Noritake Dental, Tokyo, Japan) were used in this study. Figure 1 shows schematically how the specimens were prepared. A total of 24 discs of each brand of resin composite were prepared in this study. A transparent plastic matrix strip (Hawe Striproll Transparent 10 mm, KerrHawe, Bioggio, Switzerland) was placed on the glass slide and

Table 1  Compositions of the materials used in this study

<table>
<thead>
<tr>
<th>Materials</th>
<th>Manufacturer</th>
<th>Compositions</th>
<th>Filler load</th>
</tr>
</thead>
<tbody>
<tr>
<td>Filtek™ Supreme Ultra (FSU)</td>
<td>3M, St. Paul, MN, USA</td>
<td>Non-agglomerated/non-aggregated zirconia, silica, aggregated zirconia/silica clusters, Bis-GMA, UDMA, TEGDMA, Bis-EMA, PEGDMA</td>
<td>78.5wt%/63.0vol%</td>
</tr>
<tr>
<td>Estelite Sigma Quick (ESQ)</td>
<td>Tokuyama Dental, Tokyo, Japan</td>
<td>Silica–zirconia fillers, Silica–titania fillers, Bis-GMA, TEGDMA</td>
<td>82.0wt%/71.0vol%</td>
</tr>
<tr>
<td>Beautifil II (BE2)</td>
<td>Shofu, Kyoto, Japan</td>
<td>Aluminofluoro-borosilicate glass, S-PRG filler, Bis-GMA, TEGDMA</td>
<td>83.3wt%/68.6vol%</td>
</tr>
<tr>
<td>Clearfil AP-X (APX)</td>
<td>Kuraray Noritake Dental</td>
<td>Barium glass filler, colloidal silica filler, Bis-GMA, TEGDMA</td>
<td>85.0wt%/71.0vol%</td>
</tr>
<tr>
<td>Clearfil SE One</td>
<td>Kuraray Noritake Dental</td>
<td>MDP, Bis-GMA, HEMA, hydrophobic DMA, sodium fluoride, silanated colloidal silica, accelerators, initiators, CQ, ethanol, water (pH 2.3)</td>
<td>—</td>
</tr>
<tr>
<td>Clearfil Porcelain Bond</td>
<td>Kuraray Noritake Dental</td>
<td>γ-MPTS, hydrophobic aromatic dimethacrylate</td>
<td>—</td>
</tr>
</tbody>
</table>

10-MDP: 10-Methacryloyloxydecyl dihydrogen phosphate, HEMA: 2-hydroxyethyl methacrylate, Bis-GMA: bisphenol-A-glycidyl methacrylate, TEGDMA: triethyleneglycol dimethacrylate, PEGDMA: Poly (ethylene glycol) dimethacrylate, Bis-EMA: Ethoxylated bisphenol-A-glycol dimethacrylate, UDMA: urethane dimethacrylate, MMA: methyl methacrylate, S-PRG: Surface pre-reacted glass. *The specific chemical identity and/or exact percentage (concentration) of this composition has been withheld as a trade secret.

Fig. 1  Preparation methods for composite repair microshear bond test.
a round-shaped mold (8.5 mm in diameter and 2 mm in thick) was set on it. The molds were carefully filled with each resin composite and covered with a second plastic matrix strip and compressed with a thin glass cover slip (0.1 mm thick) in order to obtain a flat surface after light curing. The resin composite was light-cured for 40 s with a halogen light-curing unit (Optilux 501, Kerr, Orange, CA, USA), which had a light output greater than 600 mW cm\(^{-2}\). The covering glass slip, and plastic strip were removed, and all the polymerized resin composite specimens were carefully removed from the molds. All the composite discs were then kept in a dark room for 30 min after which time they were stored in distilled water at 37°C for 0, 60 s, 1 week, or 1 month. We set the duration for water storage period to 1 month at the longest because it took less than 1 month for same size of composite resin specimens to fully absorb water from the pilot study (water sorption/solubility test, not shown). After each water storage period, the specimens were wiped off to remove water and air-dried using a dental three-in-one syringe for 30 s. They were divided into 3 composite surface treatment groups as follows: (Group 1) no treatment; (Group 2) a one-step self-etch adhesive, Clearfil SE One (SEO; Kuraray Noritake Dental) was applied to the resin composite resin surface, air-dried for 5 s, and then light-cured for 10 s; (Group 3) the combination of SEO and a silane coupling agent (Clearfil Porcelain Bond Activator, Kuraray Noritake Dental) was applied, air-dried for 5 s, and then light-cured for 10 s.

After all the surfaces of the specimens were treated, 6 cylinders of the resin composite were placed on the surface of the same composite resin discs using thin discs of plastic microbore Tygon tubing (Saint-Gobain Performance Plastic, Nagano, Japan) with a 0.79 mm internal diameter and 0.5 mm height. After light-curing for 40 s, all the plastic tubes were gently removed from each specimen. All of the specimens were then stored in distilled water at 37°C for 24 h.

The specimens were then subjected to the µSBS test using EZ test 500 N (Shimadzu, Kyoto, Japan) at a cross-head speed of 1 mm/min as described by Shimada et al.\(^{10}\) (n=12). The load of failure was used to calculate the µSBS in Megapascal (MPa). Subsequently, the surfaces of each fractured specimen were analyzed using a scanning electron microscope (SEM; JSM-5310, JEOL, Tokyo, Japan) at ×150 magnification to determine failure modes of the specimens. The failure modes were classified into three categories: interfacial failure (more than 70% of the failure area is between composite disc and bonding resin or additional composite), cohesive failure (more than 70% of the failure area is within the composite, or adhesive resin), or mixed failure (combination of both types of interfacial failure and cohesive failure).

The µSBS data were statistically analyzed using a three-way ANOVA (composite materials, exposure time, and surface treatments), and two-way ANOVA and t-test with Bonferroni’s correction for 2 factors (surface treatments and exposure time, and surface treatments and composites) at a significance level of 0.05.

Degree of conversion (DC)

A transparent plastic matrix strip (Hawe Striproll Transparent 10 mm, KerrHawe) was placed on the glass slab and a round-shaped mold (15 mm in diameter and 1.0 mm in thick) was set on it. The molds were slightly overfilled with one of the 4 kinds of resin composites (FSU, ESQ, BE2, and APX). Then, a second plastic matrix strip was placed on the resin composite in the mold and compressed with a thin covering glass (0.1 mm thick). The resin composite was light-cured for 40 s with a halogen light-curing unit (Optilux501, Kerr) with a light output>600 mW cm\(^{-2}\). The glass slab and plastic strips were peeled off, and all the polymerized resin composite specimens were carefully removed from the molds. The specimens were stored in the dark in air at 37°C for 24 h after the completion of exposure.

Infrared Spectroscopy (IR) spectra of micro-areas (100×100 mm) were measured in order to evaluate the quantity of remaining carbon–carbon double bonds in the specimens using a Fourier transform infrared spectrometer (JIR-100, JEOL) equipped with a microscopic unit fitted with an automatic mapping stage. The measurements were made at 0.25 mm intervals along the midline, from the irradiated surface to the base. In the IR spectra, two absorbance peaks appeared in the range, 1,600 to 1,650 cm\(^{-1}\). An appropriate baseline was drawn, and these two absorption peaks were separated into two Lorentzian curves. The intensities of absorption were determined by calculating the areas of the peaks. DC was calculated using the following equation:

\[
\text{DC} \, (\%) = 100 - \left( \frac{bc}{ad} \right) \times 100
\]

Where the intensities a–d were defined as follows: a, C=C absorbance peak at 1,637 cm\(^{-1}\) of resin pastes; b, aromatic absorbance peak at 1,608 cm\(^{-1}\) of resin pastes; c, C=C absorbance peak at 1,637 cm\(^{-1}\) of cured resins; and d, aromatic absorbance peak at 1,608 cm\(^{-1}\) of cured resins. The data were analyzed using the Statistical Package for Statistical Science (SPSS Version 15.0). One-way ANOVA was performed to analyze variance and to look for differences among the groups, followed by Tukey’s HSD (Honestly Significant Difference) post-hoc test. The significance difference was set at \(\alpha=0.05\).

### RESULTS

µSBS

There were no pretesting failures during specimen preparation for µSBS testing. Table 2 shows the mean values and standard deviations of the µSBS test. Three-way-ANOVA test revealed significant differences among the three factors, composite (\(p<0.001\)), surface treatment (\(p=0.006\)), and the duration of water storage (\(p<0.001\)). There was a significant interaction among these three factors (\(p=0.001\)). Significant interactions were found between composite materials and surface treatments for all storage times except 1 week (\(p<0.05\)), and significant interactions between surface treatments and water storage period for the all tested composites except APX (\(p<0.05\)).
Table 2  Repair microshear bond strength of newly-filled composite to resin matrix in same cured composites after water aging

<table>
<thead>
<tr>
<th>Composite surface treatments</th>
<th>Water aging of composite disk</th>
<th></th>
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<tbody>
<tr>
<td></td>
<td>None</td>
<td>60 s</td>
<td>1 week</td>
<td>1 month</td>
</tr>
<tr>
<td>No adhesive</td>
<td>33.5±2.1&lt;sup&gt;ABCab&lt;/sup&gt;</td>
<td>23.7±6.9&lt;sup&gt;ABEb&lt;/sup&gt;</td>
<td>16.4±6.3&lt;sup&gt;BDab&lt;/sup&gt;</td>
<td>13.6±6.4&lt;sup&gt;CEdbcd&lt;/sup&gt;</td>
</tr>
<tr>
<td>SE one</td>
<td>23.9±1.7&lt;sup&gt;a&lt;/sup&gt;</td>
<td>23.8±4.2&lt;sup&gt;b&lt;/sup&gt;</td>
<td>20.4±6.6&lt;sup&gt;c&lt;/sup&gt;</td>
<td>13.6±4.2&lt;sup&gt;ab&lt;/sup&gt;</td>
</tr>
<tr>
<td>SE one+Silane</td>
<td>25.4±5.5&lt;sup&gt;bd&lt;/sup&gt;</td>
<td>23.2±8.2&lt;sup&gt;f&lt;/sup&gt;</td>
<td>22.4±4.4&lt;sup&gt;a&lt;/sup&gt;</td>
<td>21.3±2.8&lt;sup&gt;bg&lt;/sup&gt;</td>
</tr>
<tr>
<td>Estelite Sigma Quick (ESQ)</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Composite surface</td>
<td>Water aging of composite disk</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>None</td>
<td>60 s</td>
<td>1 week</td>
<td>1 month</td>
</tr>
<tr>
<td>No adhesive</td>
<td>29.5±2.1&lt;sup&gt;ABCab&lt;/sup&gt;</td>
<td>17.9±5.2&lt;sup&gt;Bac&lt;/sup&gt;</td>
<td>14.7±4.3&lt;sup&gt;Bac&lt;/sup&gt;</td>
<td>14.4±4.7&lt;sup&gt;Cef&lt;/sup&gt;</td>
</tr>
<tr>
<td>SE one</td>
<td>21.8±3.7&lt;sup&gt;ab&lt;/sup&gt;</td>
<td>21.7±6.2&lt;sup&gt;e&lt;/sup&gt;</td>
<td>19.5±5.5&lt;sup&gt;a&lt;/sup&gt;</td>
<td>19.3±4.4&lt;sup&gt;ac&lt;/sup&gt;</td>
</tr>
<tr>
<td>SE one+Silane</td>
<td>19.8±4.3&lt;sup&gt;bce&lt;/sup&gt;</td>
<td>19.3±4.1&lt;sup&gt;e&lt;/sup&gt;</td>
<td>17.9±6.2&lt;sup&gt;b&lt;/sup&gt;</td>
<td>16.4±4.1&lt;sup&gt;hi&lt;/sup&gt;</td>
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<tr>
<td>Beautifil II (BE2)</td>
<td></td>
<td></td>
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<td></td>
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<tr>
<td>Composite surface</td>
<td>Water aging of composite disk</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>None</td>
<td>60 s</td>
<td>1 week</td>
<td>1 month</td>
</tr>
<tr>
<td>No adhesive</td>
<td>28.8±2.7&lt;sup&gt;A&lt;/sup&gt;</td>
<td>30.3±7.9&lt;sup&gt;Bcold&lt;/sup&gt;</td>
<td>21.5±5.7&lt;sup&gt;ABd&lt;/sup&gt;</td>
<td>23.3±4.4&lt;sup&gt;es&lt;/sup&gt;</td>
</tr>
<tr>
<td>SE one</td>
<td>27.5±6.2&lt;sup&gt;Ab&lt;/sup&gt;</td>
<td>21.4±7.9&lt;sup&gt;a&lt;/sup&gt;</td>
<td>21.2±4.7&lt;sup&gt;A&lt;/sup&gt;</td>
<td>23.7±6.9&lt;sup&gt;AB&lt;/sup&gt;</td>
</tr>
<tr>
<td>SE one+Silane</td>
<td>23.8±3.3&lt;sup&gt;f&lt;/sup&gt;</td>
<td>23.4±6.5&lt;sup&gt;Ab&lt;/sup&gt;</td>
<td>24.1±5.4&lt;sup&gt;a&lt;/sup&gt;</td>
<td>23.9±4.7&lt;sup&gt;bi&lt;/sup&gt;</td>
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<tr>
<td>Clearfil AP-X (APX)</td>
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<td></td>
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<td></td>
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<tr>
<td>Composite surface</td>
<td>Water aging of composite disk</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>None</td>
<td>60 s</td>
<td>1 week</td>
<td>1 month</td>
</tr>
<tr>
<td>No adhesive</td>
<td>31.9±6.9</td>
<td>36.3±5.0&lt;sup&gt;es&lt;/sup&gt;</td>
<td>29.0±9.8&lt;sup&gt;cd&lt;/sup&gt;</td>
<td>31.5±5.4&lt;sup&gt;ef&lt;/sup&gt;</td>
</tr>
<tr>
<td>SE one</td>
<td>27.0±8.0</td>
<td>26.9±5.8&lt;sup&gt;a&lt;/sup&gt;</td>
<td>22.8±7.4&lt;sup&gt;c&lt;/sup&gt;</td>
<td>27.0±5.8&lt;sup&gt;bc&lt;/sup&gt;</td>
</tr>
<tr>
<td>SE one+Silane</td>
<td>31.9±5.9</td>
<td>32.4±6.2&lt;sup&gt;cd&lt;/sup&gt;</td>
<td>28.8±9.3&lt;sup&gt;b&lt;/sup&gt;</td>
<td>31.4±4.4&lt;sup&gt;fg&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

Values are mean±SD. Upper-case letters refer to row. Lower-case letters refer to column. Same superscript letters indicate statistically significant differences in rows and columns (p<0.05).

In the no water-storage group, for BE2 and APX, there were no significant differences in μSBS between with and without application of one-step self-etch adhesive (p>0.05), while the adhesive application significantly decreased the μSBS of FSU and ESQ (p<0.05). The 60 s water-storage period decreased the μSBS of FSU and ESQ compared with no water-storage (FSU: p=0.01, ESQ: p=0.002), and showed no significant difference in μSBS between with and without the applications of one-step self-etch adhesive (p>0.05). In the 60 s water-storage groups without adhesive application, ESQ had the lowest μSBS among the materials. Moreover, 1-week of water-storage significantly reduced the μSBS of FSU (p=0.01), although there were no significant differences between the μSBS of FSU and ESQ after 1-week and 1-month water storage periods (p>0.05). After 1-month water storage, the adhesive application significantly increased the μSBS of FSU and ESQ (p<0.05). On the other hand, for BE2 and APX, 1-month water-storage of the repaired cured-composite did not significantly affect the μSBS (p>0.05). The specimens of APX showed the highest μSBS among the materials after 1-month water storage in each application group. In the 1-month water-storage groups, the adhesive application did not affect the μSBS of either BE2 or APX.

Adhesive application with a silane coupling agent did not significantly affect the μSBS of all the resin composites including FSU in all the experimental groups.

Failure mode analysis
Figure 2 shows the failure mode distributions of the de-bonded specimens.
Table 3  The degree of conversion (DC) of resin composites tested in this study

<table>
<thead>
<tr>
<th></th>
<th>Filtek Supreme Ultra (FSU)</th>
<th>Estelite Sigma Quick (ESQ)</th>
<th>Beautifil II (BE2)</th>
<th>Clearfil AP-X (APX)</th>
</tr>
</thead>
<tbody>
<tr>
<td>DC (%)</td>
<td>38.9±6.4\textsuperscript{a}</td>
<td>67.9±2.6\textsuperscript{b}</td>
<td>46.9±7.9\textsuperscript{c}</td>
<td>48.4±5.1\textsuperscript{c}</td>
</tr>
</tbody>
</table>

Values are mean±SD. Different superscript letters indicate statistically significant differences (p<0.05).

In the non-water storage group, non-adhesive application showed only cohesive failure in FSU and ESQ. However, the failure patterns of FSU changed from cohesive failure to interfacial failure with the application of adhesive in the non-water storage group. After 60 s water storage, half the specimens of ESQ failed at the adhesive interface, while for FSU, no specimens exhibited interfacial failure. After 1-week water-storage, the percentage of interfacial failures increased with an increase in the water storage period especially in FSU.

After 1-month of water storage of the repaired cured-composite, adhesive application reduced the number of interfacial failures among all the materials compared with non-adhesive application at each storage period. After 1-month water storage, no interfacial failures were observed in the ESQ, BE2 and APX groups with adhesive application.

DC
Table 3 shows the DC values (%) obtained from each of the resin composites. The DC values beginning with the highest first, were: ESQ; 67.9 (±2.6)\%, APX; 48.4 (±5.1)\%, BE2; 46.9 (±7.9)\%, FSU; 38.9 (±6.4)\%. ANOVA and Tukey’s HSD test revealed that there were significant differences among all the groups (p<0.05) except between BE2 and APX (p>0.05).

DISCUSSION
In this study, we investigated the influence of three experimental factors; resin composite materials, surface treatment methods and water-storage of repaired cured-composite on the repair $\mu$SBS of newly-filled composite...
applied to the resin matrix of cured-composite. Significant differences in µSBS were found among all the experimental groups. Thus, both null hypotheses tested in this study that 1) there is no difference in repair bond strength among resin composites with different DC, and that 2) water-storage does not affect the repair bond strength of resin composites with and without application of one-step self-etch adhesive, were rejected.

The existence of un-reacted C=C double bonds in the cured-composite plays a crucial role in the bonding performance of the newly-filled resin composite. The amount of un-reacted monomers in cured-composite would rely mainly on the chemical composition of the resin matrix and its polymerization behavior. Higher immediate composite-composite bond strength might occur as a result of plentiful un-reacted monomers in cured-composite. However, the un-reacted monomers in cured-composite are easily released and leached from resin matrix after aging in water, which is thought to adversely affect the composite-composite join. On the other hand, it is well-known that an oxygen-inhibited layer forms on the cured-composite, which affects the mechanical and chemical properties of the composite-composite join because un-reacted monomers in the oxygen-inhibited layer can co-polymerize with un-reacted monomers of newly-filled uncured-composite. In this study, we focused on the role of un-reacted monomers in the cured-composite on the bonding performance of the newly-filled uncured-composite to resin matrix of water-aged cured-composite. Therefore, the repaired composite surfaces were prepared and light-cured by covering with aged cured-composite. Therefore, the repaired composite newly-filled uncured-composite to resin matrix of water-storage, which is thought to adversely affect the composite-composite join. Presumably, for FSU and ESQ, the covering-method with a plastic strip on resin composite surface could not completely prevent the formation of an oxygen-inhibited layer. In the no water-storage group, the slight existence of an oxygen-inhibited layer on the cured-composite surface might produce higher µSBS without the adhesive application and ethanol/water present in Clearfil SE One could contaminate the oxygen-inhibited layer, leading to lower µSBS following adhesive application.

Sixty-second water-storage followed by air-drying reduced the µSBS of FSU and ESQ compared with no water-storage, and adhesive application had no adverse effects on their µSBS. Presumably, even if the oxygen-inhibited layer was slightly formed on the FSU and ESQ, the covering-method with a transparent plastic strip on resin composite surface would hardly be an effect of the oxygen-inhibited layer on their µSBS values in the 60 s water-storage groups because 60 s water-storage could contaminate the oxygen-inhibited layer. In the 60 s water-storage groups without adhesive application, ESQ showed the lowest µSBS among the materials. Considering the results of the failure modes, for ESQ with the highest DC (67.9%), half the specimens failed at the adhesive interface, while for FSU with the lowest DC (38.9%), there were no de-bonded specimens exhibiting interfacial failure. Additionally, after water-storage, ESQ reduced in µSBS and exhibited an increase in the number of interfacial failures, while FSU significantly reduced in µSBS with dramatic increase in the number of interfacial failures, in which there were no significant differences between the µSBS of FSU and ESQ after 1-week and 1-month of water storage. The DC value represents the quantity of carbon double bonds left unpolymerized. That is, the higher the DC value, the smaller the quantity of un-reacted monomers, which are present in the cured-composite. These results would indicate that the quantity of un-reacted monomers in the cured-composite affected immediate repair bond strength to the resin matrix of the cured-composite, and that after water storage, the un-reacted monomers in the cured-composite could not play a
significant role in repair bonding performance because the un-reacted monomers were leached during water storage\(^{24,26}\). After 1-month water-storage of the repaired cured-composite, the adhesive application of Clearfil SE One significantly increased the µSBS of FSU and ESQ with a reduced number of interfacial failures. Clearfil SE One contains the hydrophilic monomer, HEMA. Therefore, adhesive application could enhance monomer diffusion into the water-aged cured-composite with few unreacted monomers and improves surface wettability for application of the hydrophobic composite\(^{20}\).

For BE2 and APX with DC values of 46.9 and 48.4%, 1-month water-storage of the repaired cured-composite did not significantly affect µSBS. However, the number of interfacial failures increased with an increase in the water storage period. Additionally, in the 1-month water-storage group, the adhesive application did not affect the µSBS of BE2 and APX although it reduced the number of interfacial failures. These results would indicate that water-storage could slightly affect composite-composite join of BE2 and APX because the un-reacted monomers were leached. That is, after 1-month water storage of the repaired cured-composite, BE2 and APX with/without the adhesive application would have a higher quality composite-composite join than FSU and ESQ with DC of 38.9 and 67.9%. Therefore, the DC values of resin composite might not strongly affect the repair bond strengths to resin matrix of the water-aged cured-composite. The resin matrix of APX, which had stable and high µSBS even after 1-month water storage, may have an advantage for composite repair restoration. It could be speculated that various factors, such as monomer components, water uptake (water sorption and solubility), morphological and mechanical properties of the resin matrix, will affect repair bond strengths to resin matrix of the water-aged cured-composite to a greater or lesser effect. Surprisingly, in FSU and ESQ groups even after 1-month water-storage, in which most of specimens exhibited interfacial failure, µSBSs were not zero MPa. Further research is indicated to evaluate the repair bond strength with different DC of each resin composite to explore the correlation between DC and µSBS, and determine adhesion to the resin matrix of cured-composite after long-term aging, simulating clinical situations.

**CONCLUSIONS**

The quantity of un-reacted monomers in the repaired cured-composite contributed to the bonding performance of newly-filled uncured-composite to the resin matrix of cured-composite without water storage, and the application of a one-step self-etch adhesive could not improve their repair bonding performances. Following 1-month water storage of the repaired cured-composite, FSU and ESQ exhibited reduced repair bonding performance to the resin matrix of the cured-composite, but adhesive application could improve their repair bond strengths. On the other hand, BE2 and APX did not exhibit significantly reduced repair bond strengths to resin matrix of the water-aged cured-composite, but the quality of the composite-composite join slightly deteriorated, in which the adhesive application did not increase repair bond strengths. For water-aged cured-composite resins, the repair bond strength was no longer affected by the amount of initially residual un-reacted resin monomers.

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