Evaluation of marginal failures of dental composite restorations by acoustic emission analysis

Ja-Uk GU¹ and Nak-Sam CHOI²

¹ Department of Mechanical Engineering, Graduate School, Hanyang University, 17 Haengdang-dong, Seongdong-gu, Seoul 133-791, Korea
² Department of Mechanical Engineering, Hanyang University, 1271 Sa 3-dong, Sangrok-gu, Ansan-si, Gyeonggi-do 426-791, Korea

Corresponding author, Nak-Sam CHOI; E-mail: nschoi@hanyang.ac.kr

In this study, a nondestructive method based on acoustic emission (AE) analysis was developed to evaluate the marginal failure states of dental composite restorations. Three types of ring-shaped substrates, which were modeled after a Class I cavity, were prepared from polymethyl methacrylate, stainless steel, and human molar teeth. A bonding agent and a composite resin were applied to the ring-shaped substrates and cured by light exposure. At each time-interval measurement, the tooth substrate presented a higher number of AE hits than polymethyl methacrylate and steel substrates. Marginal disintegration estimations derived from cumulative AE hits and cumulative AE energy parameters showed that a significant portion of marginal gap formation was already realized within 1 min at the initial light-curing stage. Estimation based on cumulative AE energy gave a higher level of marginal failure than that based on AE hits. It was concluded that the AE analysis method developed in this study was a viable approach in predicting the clinical survival of dental composite restorations efficiently within a short test period.

Keywords: Composite resin, Acoustic emission, Contraction stress, Marginal failure, Dental restoration

INTRODUCTION

Polymerization shrinkage occurs during light-curing of resin-based restorative materials inside a cavity. When polymerization shrinkage takes place under confinement—due to bonding of a composite resin to cavity wall, it induces contraction stresses at the composite-tooth bond and in surrounding tooth structure. Generated stresses can cause cuspal deflection, enamel crack propagation, debonding, and marginal gap formation at tooth-restoration interface. Polymeric shrinkage stress is affected by several dominant factors: tooth region, cavity configuration factor (C-factor, defined as the ratio of a restoration’s bonded to unbonded surfaces), restorative material composition, restoration method (incremental placement versus bulk filling technique), and treatment conditions (e.g., surface pretreatment, light curing conditions). Several alternatives and techniques were recommended to improve these factors to reduce contraction stress generation. One means to evaluate the effectiveness and viability of these recommendations is to examine the marginal integrity of dental composite restorations. Optical microscopy, scanning electron microscopy (SEM), dye penetration testing, and nondestructive X-ray micro-computed tomography (micro-CT) have been used to observe the marginal quality of the light-activated surfaces of specimens or sectioned surfaces.

Lately, acoustic emission (AE) analysis is employed for nondestructive examination of interfacial debonding, caused by polymerization shrinkage stress, in composite resin restorations. This nondestructive testing technique is able to monitor, in real time, defect formation and failure within a material through the detection and analysis of AE signals, which are stress waves rapidly released from strain energy stored in the material. For example, AE hits and amplitudes were used to elucidate the microfracture mechanisms of dental composite resins.

In our previous works, AE signals were monitored in real time to observe the marginal disintegration process taking place in the interior of restored tooth specimens. Results showed that AE events could be used as a nondestructive evaluation index for marginal failures caused by polymerization contraction stress. In separate studies by Li et al. and Liu et al., monitoring of AE events revealed that the higher the shrinkage stress, the more likely it was for interfacial debonding to occur in composite resin restorations during curing.

In this study, a simple and easy-to-use AE analysis method was developed for nondestructive monitoring, in real time, the marginal failure process of dental composite restorations. An AE sensor was directly attached to the substrate using a mechanical fixture. AE signals were recorded as a function of time to monitor marginal failure development in dental composite restorations. The viability of this novel AE measurement method would be assessed by the effectiveness of AE parameters—in terms of hits, amplitude and energy—in evaluating the marginal failure states of dental composite restorations.
MATERIALS AND METHODS

Polymerization contraction stress evaluation
1. Preparation of penetrated ring-shaped substrates
Penetrated (i.e., hollow) ring-shaped substrates of 6 mm inner diameter, 8 mm outer diameter, and 2 mm height were modeled after a Class 1 cavity with a C-factor of 2.33. Substrates were prepared using three kinds of materials: polymethyl methacrylate (PMMA), stainless steel (SUS 304), and human molar teeth. PMMA was selected for this study because it is a transparent and colorless polymer commonly used as a denture base material. Stainless steel was selected because it is a commonly used prosthetic material to restore primary or permanent teeth.

For PMMA and stainless steel substrates, they were produced through a careful process of machining, drilling, and grinding. For the human molar teeth, they were immersed in a 0.9% NaCl solution at 2°C after extraction.

2. Preparation of composite restorations in penetrated substrates
A light-cure, self-etch dentin bonding agent (Clearfil S3, Kuraray, Japan) was applied to the inner hollow surfaces of the penetrated substrates following the manufacturer’s instructions. After gentle air-blowing, the adhesive layer was cured for 10 s using a light-emitting diode (LED) curing light (Pencure, J. Morita, Japan; 1,000 mW/cm²). The LED apparatus is easy and convenient for clinical use, and was hence used in this study to obtain results representative of the clinical setting.

Using a dental hand tool, a composite resin (Clearfil AP-X, Kuraray, Japan) containing Bis-GMA (2,2-bis[4-(2-hydroxy-3-methacryloxy-propoxy)-phenyl]propane) matrix and barium glass filler particles was tightly packed into the hollow space of each substrate. The packed composite resin was cured by exposure to LED light for 20 s. The LED light was maintained at a 3-mm distance from and oriented parallel to the top surface of the substrate.

For the human molar tooth substrate, its outer surface was wrapped with a wet tissue paper after LED light exposure. This was done to prevent tooth fracture caused by prolonged dehydration during the experiment.

3. Contraction stress measurement
Using the elastic ring method as previously described, polymerization contraction stress developed at composite restoration margin (i.e., composite-substrate interface on the inner surface of ring substrate) was measured based on circumferential stress on the outer surface of ring substrate. Measurements were performed seven times for each ring substrate.

The elastic cylindrical shell model was applied to calculate contraction stress as a function of time. Young’s moduli (E) were assumed as follows for a mixed ratio of 1:1 of dentin and enamel layers: \( E_{\text{PMMA}} = 3.1 \text{ GPa} \), \( E_{\text{SUS 304}} = 202 \text{ GPa} \), and \( E_{\text{tooth}} = 49 \text{ GPa} \). It must be highlighted that the thus-obtained contraction stress at composite restoration margin was not a true shrinkage stress generated within the composite resin based on homogeneous and adequate bonding of composite resin and ring substrate. This value was only an ‘apparent’ shrinkage stress based on these assumptions: imperfect composite-substrate interface and interfacial cracking of composite.

Acoustic emission analysis
1. Preparation of non-penetrated ring-shaped substrates and composite restorations
Non-penetrated, cavity-type, ring-shaped substrates of 6 mm inner diameter, 8 mm outer diameter, 2 mm inner depth, and 3 mm outer height were modeled after a Class 1 cavity with a C-factor of 2.33. Like the penetrated ring-shaped substrates used for contraction stress measurement, these non-penetrated substrates were also prepared using three kinds of materials: PMMA, stainless steel (SUS 304), and human molar teeth. Procedures and methods used to prepare the non-penetrated substrates for AE measurement was as per those described for the penetrated substrates used for contraction stress measurement.

To prepare the composite restorations, the materials and experimental conditions used for these non-penetrated, cavity-type substrates were also as per those described above for the penetrated, hollow substrates.

2. AE measurement
Using a mechanical fixture, an AE sensor (Micro30, Physical Acoustics Corp., USA) —of 9.5 mm diameter and with a detectable frequency range of 100–600 kHz and peak sensitivity of 250 kHz— was directly attached to the non-penetrated substrate (Fig. 1). A vacuum grease was used as the acoustic coupling agent, and pressure was applied by tightening a mechanical screw (Fig. 1b). Background noises such as friction and impacts, arising from surface abrasion and/or slip between hard materials at contact sites, were eliminated by inserting a soft, thin rubber sheet between the substrate and fixture.

AE hits, amplitudes, and energy were measured as a function of time under these settings: 40 dB preamplifier, 25 dB threshold level, and 4 MHz sampling rate. AE signals were recorded in real time for 90 min from the start of LED light irradiation on the packed composite resin in the substrate. Recorded data were written to a computer hard disk in an AE detection system (MISTRAS 2001, Physical Acoustics Corp., USA).

Marginal disintegration measurement
After LED light curing of composite resins, 0.5-mm-thick slices were sectioned from the top surfaces of non-penetrated ring substrate specimens. After polishing, composite restoration margins were analyzed by scanning electron microscopy (SEM) to measure maximum gap width and gap percentage (GP). GP was calculated as the summation of gap lengths on the inner
circumference of the ring substrate.

Statistical analysis
For multiple comparisons, acquired data were analyzed by Kruskal-Wallis test. Data were presented as mean±standard error. Differences of $p<0.05$ were considered statistically significant.

RESULTS

Polymerization contraction stress of composite restorations
As the composite resin changed from a gel state to a solid state during the 20 s of LED light exposure, the polymerization contraction rates of composite restorations became rapid (Fig. 2). Contraction rates were highest at around 28 s. After which, the resin cured itself at a slow contraction rate.

For steel and human molar tooth, each material displayed two different sets of stress behaviors in the period after reaching maximum stress. However, the dominant behavior of both materials was that contraction stress considerably decreased with increasing time. In a very small number of cases, composite restorations in some steel and human molar substrates displayed a slow increase in contraction stress until the end of the 90-min test. For PMMA, the composite restorations showed a slight increase in contraction stress until the end of test.

Table 1 lists the the maximum stress and maximum contraction rate data. Stainless steel displayed the highest maximum contraction rate which was about 8.3 times higher than that of PMMA. Such a large value for the stiff steel material was caused by its high Young’s modulus as compared to low modulus of PMMA ring\(^{9}\). For the human molar tooth, its maximum stress and maximum contraction rate were ranked intermediate between and statistically different from stain steel and PMMA. The time at which the human molar tooth reached its maximum contraction rate was 28.3±2.28 s, which was not significantly different from steel and PMMA substrates ($p>0.05$).

Table 1  Maximum contraction stress levels and maximum contraction rates according to substrate material

<table>
<thead>
<tr>
<th>Substrate</th>
<th>Maximum contraction rate (MPa/s)</th>
<th>Time of maximum contraction rate (s)</th>
<th>Maximum contraction stress $\sigma_{\text{max}}$ (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PMMA</td>
<td>0.03±0.002</td>
<td>29.2±2.59</td>
<td>1.15±0.08</td>
</tr>
<tr>
<td>Stainless steel</td>
<td>0.25±0.03</td>
<td>26.9±1.56</td>
<td>11.26±1.43</td>
</tr>
<tr>
<td>Human molar tooth</td>
<td>0.09±0.002</td>
<td>28.3±2.28</td>
<td>5.44±0.73</td>
</tr>
</tbody>
</table>

![Fig. 1](image1.png) Schematic diagram (a) and photo (b) of acoustic emission measurement with an AE sensor directly attached to the substrate during LED light curing of composite resin.

![Fig. 2](image2.png) Polymerization contraction behaviors at composite restoration margins measured from the inner surfaces of ring substrates.
Marginal failure of composite restorations
During composite resin curing, large tensile stress propagated cracks along the inner surface of the substrate. These cracks grew to become marginal gaps (Fig. 3).

The marginal gap widths of human molar tooth were statistically smaller than those of steel, but significantly larger than those of PMMA. GP and maximum gap width measured in this study were: 38.4±4.5% and 2.7±0.3 µm for human molar; 72.1±3.4%, 5.2±0.5 µm for stainless steel; and 3.2±0.5% and 1.1±0.2 µm for PMMA.

Acoustic emission behavior of composite restorations
Figure 4 shows the AE hits detected from the non-penetrated, cavity-type substrates during the 90-min monitoring time from the start of composite resin curing. At some time intervals, where the mean number of AE hits was lower than ‘1’ on the chart, it meant that no AE hits were detected at that time interval.

For PMMA, a very small number of AE hits was detected, and that the majority of the detected AE events were concentrated at the initial stage of composite curing (Fig. 4). Cumulative AE hits (CH) of PMMA showed a steep increase during the initial curing period, followed by a very slow increase and then almost reaching a plateau. The low CH value of PMMA indicated good marginal integrity and agreed with the marginal gap width results obtained by SEM above. Between human molar tooth and stainless steel, the former displayed a markedly higher CH than the latter (Fig. 4). For the three types of substrate materials, about 67% of their cumulative AE hits were detected during the first 5 min of the entire monitoring period.

Figure 5 shows the average amount of AE energy released at each time interval by composite restorations in each type of substrate material. Cumulative AE energy, CE, for each substrate material was calculated using the formula below:

\[
CE = \sum_{i=1}^{CH} V_i^2
\]

where \(V_i\) is the voltage amplitude (mV) of each AE hit.
DISCUSSION

In this study, AE signals detected during the curing of composite restorations were nondestructive means of monitoring cracks in composite resins and marginal gaps at composite-substrate interfaces —which were otherwise unobservable from the outside of the specimens. Through this study, we sought to identify the AE parameters which could provide a viable evaluation of marginal failure states in composite resin restorations.

Polymerization contraction behavior of composite restorations

Polymerization contraction stress rose rapidly during the 20-s LED light exposure, with the contraction rate peaking at 26–30 s for all substrate materials (Fig. 2 and Table 1). This high contraction rate during the initial curing stage most probably damaged the integrity of the composite-substrate interface because of two reasons. First, although the main composite resin body was rapidly solidifying because it was fully irradiated by direct exposure to LED light, composite resin at restoration margin near the adhesive layer remained in a delayed semi-solidified state. Second, contraction rate remained high throughout the initial 1 min. During this time interval, stress level at restoration margin already reached 60–80% of the maximum stress level ($\sigma_{\text{max}}$). After the initial 1 min, contraction stress reached the $\sigma_{\text{max}}$ point at a slow contraction rate.

After contraction stress reached the $\sigma_{\text{max}}$ point, there seemed to be no further increase in stress level for PMMA (Fig. 2). For stainless steel and human molar tooth, there was a significant decrease in stress level caused by the initiation and growth of marginal defects in composite restorations.

Results from Fig. 2 and Table 1 of this study showed that a slew of factors affected the maximum stress level, maximum contraction rate, and time of maximum contraction rate of composite restorations: irradiance conditions, exposure time, restorative material composition, and C-factor. To maintain good marginal integrity between a composite resin restoration and the cavity walls, a composite resin characterized by low polymerization contraction stress level and low contraction rate, especially during the early curing stage, would be highly recommended and preferred.

Marginal failure of composite restorations

Although bonding procedure was carried out as per the manufacturer’s instructions in this study, polymerization shrinkage damaged the bond quality. Polymerization contraction stress induced interfacial cracking of the substrate along the inner surface. For PMMA, marginal gap formation was very low because of very low contraction stress (Fig. 2). For human molar dentin (Fig. 3), marginal gap formation was statistically lower than that of steel.

GP and maximum gap width, which characterized the extent of marginal failure of composite restorations, were obtained in this descending order: stainless steel>human molar tooth>PMMA. Maximum stress level and maximum contraction rate, as explained above, accounted for this result. Therefore, a high contraction stress level would induce large gap formation.

Acoustic emission behavior of composite restorations

Blast-type AE signals due to marginal disintegration had a dominant frequency band of 100–290 kHz, which represented some transient release of strain energy stored at the marginal region. Amplitude distribution covered the range of 25 dB to 58 dB, irrespective of substrate material. Statistical analysis by Kruskal-Wallis test revealed that there were no significant differences in amplitude data among the three substrate materials ($p>0.05$).

In a study by Liu et al. which employed different C-factors, it was shown that the higher the shrinkage (contraction) stress at tooth-restoration interface, the more AE hits were detected. Results of this study showed that besides the effect of contraction stress level, AE hits and AE energy also depended on the substrate material, restorative material composition, and bonding quality.

For all the three substrate materials, CH and CE showed a steep increase during the initial curing period, only to be followed by a very slow increase until the end of the 90-min monitoring period (Figs. 4 and 5). For PMMA, only a very small number of AE hits was generated, thus indicating good marginal integrity. For the human molar, its average number of AE hits during the initial curing period was significantly higher than that of steel. Indeed, the CH and CE values of human molar at the end of 90-min monitoring period were the highest among the three substrate materials. Interestingly, the maximum contraction stress level of human molar tooth was 52% lower than that of steel (Table 1). These seemingly contradictory results could be explained by the weak bond strength between dentin and composite resin.

AE hits detected during the light-curing of composite resin restorations might not originate from interfacial and composite resin defects only, but also from fractures in dentin substrate. Another factor which might influence AE signal characteristics were the mechanical properties of heterogeneous composite resin and adhesive layer, which could vary with the curing reaction. Therefore, AE source characterization needs to be studied further using different frequencies and intensities of AE signals. Studies have shown that discrimination of AE signals using amplitude distribution, pattern recognition and peak frequency analyses could achieve the classification of several failure modes in fiber-reinforced composite and metal laminates.

Marginal failure evaluation using AE parameters

Under the assumption of a linear relationship, the state of marginal disintegration (MD) of a dental composite restoration as a function of measurement time could be estimated using the following equations based on CH and CE:
Table 2  Estimations of marginal disintegration (MD) states of dental composite restorations as a function of time using AE parameters of cumulative hits (CH) and cumulative energy (CE)

<table>
<thead>
<tr>
<th>Substrate</th>
<th>AE parameter</th>
<th>Marginal disintegration state (%)</th>
<th>Damage maturity (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Initial 1 min</td>
<td>Initial 5 min</td>
</tr>
<tr>
<td>PMMA</td>
<td>CH</td>
<td>2.6±0.2</td>
<td>2.9±0.1</td>
</tr>
<tr>
<td></td>
<td>CE</td>
<td>2.6±0.3</td>
<td>2.9±0.2</td>
</tr>
<tr>
<td>Stainless steel</td>
<td>CH</td>
<td>52.6±6.7</td>
<td>57.9±5.2</td>
</tr>
<tr>
<td></td>
<td>CE</td>
<td>62.1±8.5</td>
<td>65.7±6.7</td>
</tr>
<tr>
<td>Tooth (dentin)</td>
<td>CH</td>
<td>20.6±5.1</td>
<td>25.9±6.0</td>
</tr>
<tr>
<td></td>
<td>CE</td>
<td>26.9±5.9</td>
<td>34.1±6.4</td>
</tr>
</tbody>
</table>

\[ MD_h(t) = \frac{CH(t)}{CH_{total}} \cdot GP(\%) \quad (2) \]

\[ MD_e(t) = \frac{CE(t)}{CE_{initial}} \cdot GP(\%) \quad (3) \]

where \( CH(t) \) and \( CE(t) \) are values of CH and CE respectively at measurement time \( t \).

Table 2 shows the estimated MD states of dental composite restorations according to substrate material and at different measurement time intervals. Estimations using the AE parameters of CH and CE showed significant marginal disintegration within the initial 1 min, because majority (54–86%) of the final MD state at 90 min was realized within the first minute for all the three substrate materials.

Based on Equation (1), CE took into account the effects of both AE hits and amplitudes. When compared to the MD states based on CH, CE-based MD states showed higher levels of marginal gap formation within the initial 1 min.

Leveraging on the estimated MD data in Table 2, a damage maturity index \( (m) \) as a function of measurement time \( t \) was calculated using the equation below:

\[ m = \frac{MD(t)}{MD_{total}} \quad (4) \]

where \( MD(t) \) was the MD value at measurement time \( t \) and \( MD_{total} \) was the MD value at the end of 90-min monitoring period.

For all the three substrate materials, the values of \( m \) were higher than 0.54 at 1-min interval and higher than 0.67 at 5-min interval. When based on CH, the human molar tooth yielded the lowest \( m \) value while PMMA yielded the highest. Therefore, the value of \( m \) depended on substrate material.

When based on CE, the \( m \) values of stainless steel and human molar tooth substrates were significantly higher by at least 11% than those based on CH (Table 2). The CE parameter allowed a high damage maturity estimation to be yielded within a short test period.

Based on Equation (4), \( MD(t) \) could be obtained by taking only 1 min of the AE test. With \( m \) values already available prior to \( MD(t) \) calculation, the final MD state could be efficiently predicted for a clinical application using Equation (4) within a short time.

**CONCLUSIONS**

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

1. Acoustic emission (AE) analysis was a nondestructive method to evaluate the marginal disintegration (MD) states of dental composite restorations during light-curing.
2. AE hits indicated the initiation and propagation of marginal gaps in composite restorations, and the cumulative AE hits (CH) generated by human molar tooth was higher than those generated by PMMA and stainless steel.
3. Compared to CH, the cumulative AE energy (CE) parameter gave a higher level of marginal gap formation within an initial 1 min of AE test.
4. Damage maturity index, which could be derived from CH or CE, gave a useful indication of the state of marginal disintegration at a particular measurement time interval, making the AE analysis method an efficient means of evaluating marginal failure states in dental composite restorations.

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