Filler size of resin-composites, percentage of voids and fracture toughness: is there a correlation?

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The objective of this study was to investigate the correlation between filler size, fracture toughness and voids. Seven model resin composites and one commercial have been used in the study. A single edge notch mould was used to prepare samples (n=8). A selected area of 1mm below and above the notch was scanned with micro CT and then the percentage of voids calculated. A universal testing machine was used to measure fracture toughness. Percentage of voids and fracture toughness data were analysed using ANOVA and post hoc methods were performed to check any significant differences between materials tested (p<0.05). Conclusion: Filler size is strongly correlated to % voids but has no effect on fracture toughness.

Keywords: Resin composite, Voids, Fracture toughness, Porosity, Micro-CT

INTRODUCTION

High demands for aesthetic dentistry and new developments in resin-composites have led many dentists to use resin-composite instead of amalgam for restoration of teeth including posterior teeth. The success rate of direct resin-composite restorations in posterior teeth has been found to be 90% over 5 years12,13, dropping to 77% at 8 years14. Despite the good clinical success, direct composite restorations have been associated with undesirable characteristics such as excessive wear, marginal leakage caused by polymerization shrinkage, voids, sensitivity after placement, and insufficient proximal contact and contour15. The two main reasons for replacement of resin-composite restorations are secondary caries and fracture16; these are related to resin-composite being a brittle material and its clinical longevity is affected by surface flaws, which may propagate through the material matrix leading to fracture and subsequent caries17. Resin composites with higher fracture toughness will better withstand high stress levels18 and are thus have improved clinical service.

Another major disadvantage of direct composite is the presence of voids within the final restoration which may arise due to manufacturing procedures or handling techniques9. When present, these voids may cause fractures within a restoration such as9: marginal leakage and discoloration, increased wear due to the stress concentration around voids, decreased flexural strength and also incomplete adhesion between the resin-composite and dentine11.

There have been several attempts to solve the problems associated with direct composite restorations, including increasing the percentage of filler content in the composite matrix and reducing the size of filler particles12-14. Several studies showed that heavier filler loading would result in increase in fracture toughness of the material15, while others concluded that filler content has no role in fracture behaviour16. Generally the filler contents have a significant influence in the mechanical properties, with the highly filled composites being the strongest17. The aim of this study is to investigate the effect of filler size on fracture toughness and presence of voids. The objectives were to measure fracture toughness KIC on a series of model resin-composites and one commercial using the Single-Edge Notch technique and to use microtomography to quantify the voids in the same materials. The model composites used in the study had different filler distributions (unimodal, bimodal and trimodal) while the commercial composite used was a multimodal. The filler shape of all composites used was irregular, spherical or combination as in the commercial composite. The hypotheses of this study were that different filler size has no effect on i) the fracture toughness of resin-composites; and, ii) the presence of voids within resin-composites.

MATERIALS AND METHODS

Sample preparations

Single-edged notch (SEN) specimens (n=8) for each group, conforming to British Standard 5447 (1977), were prepared in a PTFE-lined brass mould which could be split so that no force was required to remove the set specimen from the mould. The specimen size and geometry are shown in Fig. 1. The overall external dimensions were 3 mm×6 mm×25 mm, and a sharp notch (a) to half the beam height (w) was 3 mm. A central sharp notch of specific length was produced by inserting a straight-edged scalpel blade into an accurately fabricated slot at mid-height in the plastic mould which extended down half the height to give a/w=0.5. The blade had a straight cutting edge, honed on both sides with a blade edge radius less than 0.3 μm. The crack plane was perpendicular to the specimen length. Samples consisted of 7 light cured model composites (Ivoclar Vivadent, Schaan, Liechtenstein) together with an established commercially available formulation (Tetric
Ceram-Ivoclar Vivadent, Schaan, Liechtenstein) used as a control. Table 1 illustrates the composition of materials used. They were all polymerised with a QTH light curing unit (Optilux 501, Kerr SDS, Peterborough, UK) with a 10 mm diameter curing tip and an irradiance of 550 mW/cm² as measured with the radiometer incorporated into the appliance. To ensure optimal curing depth each sample was cured from the top surface for 60 s and then from each side for further 60 s after disassembling of the mould.

Table 1  Composition of resin-composites used in the study including filler size, shape, content and distribution. The resin matrix was the same in all materials

<table>
<thead>
<tr>
<th>Resin-Composite</th>
<th>Filler Particles (Ground Glass [Ba-Al-B-silicate glass])</th>
<th>Matrix</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Shape</td>
<td>Size (nm)</td>
</tr>
<tr>
<td>I1</td>
<td>Irregular</td>
<td>450</td>
</tr>
<tr>
<td>I2</td>
<td>Irregular</td>
<td>700</td>
</tr>
<tr>
<td>I3</td>
<td>Irregular</td>
<td>1,000</td>
</tr>
<tr>
<td>I4</td>
<td>Irregular</td>
<td>1,500</td>
</tr>
<tr>
<td>I5</td>
<td>Irregular</td>
<td>450, 1,000(1:3)</td>
</tr>
<tr>
<td>I6</td>
<td>Irregular</td>
<td>450, 700 &amp;1,500 (1:1:3)</td>
</tr>
<tr>
<td>SP</td>
<td>Spherical</td>
<td>100</td>
</tr>
<tr>
<td>TC Lot : F53738</td>
<td>Irregular &amp; Spherical</td>
<td>40, 200 &amp;1,000</td>
</tr>
</tbody>
</table>

Microtomography 3-D Scanning
All samples were scanned with a high resolution micro-CT (Model 1072, SkyScan, Kontich, Belgium) operated under the following conditions: 98 μA fixed current; voltage of 100 kV voltage; 19 μm pixel size at 14.39×14.39 resolution; 180° rotation; 4 s exposure time and averaging by one frame. Data obtained were then input into a software package (N-Recon, SkyScan) and reconstructed resulting in 100 slices of 2D images. Each 2D image, using CTAn software (SkyScan), was then converted into a 3D image. The percentage of volume occupied by voids within each sample was calculated in an area 1.0 mm below and 1.0 mm above the notch.

Fracture toughness
Fracture toughness of each sample was tested using a universal testing machine (Zwick/Roell Z020, Leominster, UK) at cross head speed of (0.127 mm/min). The maximum force at fracture (P) was recorded and fracture toughness (Ktc) was calculated using the following equation:

\[ K_{tc} = \frac{3PL}{BW^{3/2}} \cdot Y \]

Where P=peak load at fracture; L=length; B=width; W=height; Y=calibration functions for given geometry.
\[ Y = [1.93(a/w)^{1/2} - 3.07(a/w)^{3/2} + 14.53(a/w)^{5/2} - 25.11(a/w)^{7/2} + 25.80(a/w)^{9/2}] \]

**Data analysis**

Voids and \( K_{IC} \) data for the eight groups were analysed with a statistical software package (SPSS ver 16.0, IL, USA) using One-Way ANOVA (\( p<0.05 \)). Prior to post-hoc tests, data were analysed for equal variances using Levene’s test for homogeneity (\( p<0.05 \)): for \( K_{IC} \) data, equal variances could be assumed thus Bonferroni test was applied, however Dunnett’s T3 was applied for data of voids as equal variances could not be assumed.

**RESULTS**

The voids could be seen in both 2D and 3D images (Fig. 2). The percentages of voids for each group are shown in Table 2. TC (40: 200: 1,000 nm) exhibited the lowest percentage of voids amongst all the eight groups (0.27%). For the unimodal composites the percentage of voids ranged from 0.44% for SP (100 nm) to 3.53% for I4 (1,500 nm). Generally the percentage of voids increased with the increase in filler size \((r=0.97)\) (Fig. 3). The difference of the %voids between the materials was statistically significant \((p<0.05)\).

For unimodal composites the \( K_{IC} \) values ranged between 1.50 MNm\(^{-1}\) to 1.10 MNm\(^{-1}\), with the highest \( K_{IC} \) value seen with I1 (450 nm). Overall, the highest \( K_{IC} \) value was seen with TC (40: 200: 1,000 nm). Generally the filler size had no effect on the fracture toughness \((r=0.02)\). Statistically significant differences were present between some of the materials tested \((p<0.05)\).

**DISCUSSION**

It has been shown that voids have a negative effect on the physical and mechanical properties of resin-composite material such as: leakage around the margin and subsequent discolouration, increased wear and reduced flexural strength\(^{10}\). Voids have been investigated and measured with different techniques, such as light and electron microscopes both of which are destructive methods\(^{9,10,18,19}\), and the novel non destructive method using micro CT\(^{20,21}\).

In this study, filler size was found to have an effect on the presence of voids. As filler size increases so does the percentage of voids with the highest percentage of 3.53% found in a resin-composite material which contained the largest filler particles of 1,500 nm. It is clear that increasing the filler size increases voids and this is obvious from the strong correlation between %Voids and filler size \((r=0.97)\). Hence the first null hypothesis was rejected.

The presence of voids within resin-composite material could be due to manufacturer error or poor handling technique\(^{9}\). These two factors will contribute to the origin of voids. The relation of viscosity of resin-composite (flowable-composite) and voids has also been studied and it has been shown that flowable-composite reduce voids within class II restoration\(^{10}\). In a recent study the effect of filler size and temperature on handling properties of resin-composite in terms of packing stress and viscosity has been investigated and it has been shown that as filler size and temperature increase packing stress and viscosity also increase\(^{22}\). Since packing stress and viscosity will affect the handling properties it can be assumed that as filler size increased the handling technique was compromised. Therefore, one could extrapolate that these composites with larger
Filler sizes could result in increased voids. The present study confirms that assumption.

Fracture is one mode of clinical failure of resin composite restoration; it could be bulk or marginal fracture. The resistance of a material to fracture is measured by fracture toughness ($K_{IC}$) which is an intrinsic property of a resin-composite material and its resistance to crack propagation, and a material which has a higher $K_{IC}$ value has the ability to resist the initiation and propagation of the crack.

Unlike %Void data, $K_{IC}$ values are not affected by variations in filler size or distribution ($r=0.02$). Hence

### Table 2  Mean (SD) values of Voids % and $K_{IC}$ (MNm$^{-1.5}$) of all resin composites tested

<table>
<thead>
<tr>
<th>Groups</th>
<th>Voids % Mean(SD)</th>
<th>$K_{IC}$ (MNm$^{-1.5}$) Mean(SD)</th>
</tr>
</thead>
<tbody>
<tr>
<td>I1  (450 nm)</td>
<td>0.59 (0.05)</td>
<td>1.50 (0.11)</td>
</tr>
<tr>
<td>I2  (700 nm)</td>
<td>1.49 (0.23)</td>
<td>1.33 (0.04)</td>
</tr>
<tr>
<td>I3  (1000 nm)</td>
<td>2.96 (0.12)</td>
<td>1.20 (0.08)</td>
</tr>
<tr>
<td>I4  (1500 nm)</td>
<td>3.53 (0.24)</td>
<td>1.26 (0.07)</td>
</tr>
<tr>
<td>I5  (450:1000 nm)</td>
<td>1.18 (0.28)</td>
<td>1.27 (0.06)</td>
</tr>
<tr>
<td>I6  (450:700:1000 nm)</td>
<td>0.56 (0.09)</td>
<td>1.45 (0.06)</td>
</tr>
<tr>
<td>TC  (40:200:1000 nm)</td>
<td>0.27 (0.07)</td>
<td>2.00 (0.09)</td>
</tr>
<tr>
<td>SP  (100 nm)</td>
<td>0.44 (0.04)</td>
<td>1.10 (0.06)</td>
</tr>
</tbody>
</table>

Within each column; different superscript letters indicate significant differences between the groups ($p<0.05$).

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Fig. 3  Bar chart illustrating Mean and SD of Voids% for all materials with linear correlation shown between unimodal composites and Voids% ($r=0.97$).

Fig. 4  Bar chart illustrating Mean and SD of Fracture toughness for all materials.
the second null hypothesis was confirmed. Furthermore, there was no correlation between $K_{IC}$ and voids ($r=0.20$). Tetric Ceram (40:200:1000) had the highest value which probably could be to higher filler loading. The lowest value was obtained by SP that has the smallest fillers (100 nm) and is the only one with solely spherical shape fillers. Additionally, it has the lowest filler content, the combination of these factors seem to result in inferior mechanical properties.

Filler size has previously been investigated in relation to fracture toughness: one study suggests that resin-composite material with 80% wt of fillers, 10% of which is microfiller, would have an optimum fracture toughness, whereas another study showed $K_{IC}$ values of hybrid and nanofilled composites are significantly higher than those of microfilled composites.

Generally there is a lack of published data regarding the relationship between fracture toughness and presence of voids. Studies relating to polymers have shown that fracture toughness decreases markedly with the increase of voids. Voids also significantly reduced the fracture toughness of Bis-GMA based composite with hydroxyapatite whiskers.

The use of model composite has proved to be very useful in elucidating trends between properties. In this paper it was shown that the size of filler particles within resin-composites is directly related to percentage of voids but does not influence fracture toughness.

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REFERENCES