Original Paper

Multi-scale analysis of the influence of filler shapes on the mechanical performance of resin composites using high resolution nano-CT images

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Conflict of interest: None

Highlights

- Fracture criteria of resin composites (RCs) were assessed at the micro-scale.
- Homogenization analysis confirmed anisotropy of the RCs at the micro-scale.
- Maximum principal strain was identified as a useful fracture criterion of the RCs.
- Localization analysis was used to visualize RC micro-scale strain concentration.
- Multi-scale analysis confirmed the advantage of irregular-shaped filler.

ABSTRACT

Objective: This aim of this study was to investigate the criteria for predicting the fracture initiation of resin composites (RCs) at the micro-scale and assess the influence of filler shapes on the flexural properties of RCs by combining nano-CT imaging and *in silico* multi-scale analysis.

Methods: Experimental RCs composed of irregular-shaped (IS) silica filler (31.2 vol%/50.0 wt%) and Bis-GMA/TEGDMA were prepared. The RC specimens were scanned by a nano-CT with 500-nm resolution, and 10 micro-scale models (100 × 100 × 100 μm) were randomly extracted from a scanned region. *In silico* micro-scale models containing sphere-shaped (SS) fillers with the same volume content as the experimental RC were designed. Each RC model's elastic modulus and Poisson's ratio at the macro-scale were calculated using homogenization analysis. The flexural strength of the RC models were predicted by finite element analysis using the elastic moduli and Poisson's ratio values.

Results: Significantly greater elastic modulus values were obtained in the x, y, and z directions for RC models containing IS fillers than SS fillers. Similarly, smaller Poisson's ratio values were observed in the y and z directions for RC model containing IS fillers than SS fillers ($p<0.05$). The flexural strength of RC model containing IS fillers was significantly greater than the RC model containing SS fillers ($p<0.05$).

Significance: The *in silico* multi-scale analysis established in this study demonstrated that RC model containing irregular-shaped fillers had greater flexural strength than RC model loaded with SS fillers, suggesting that the mechanical strength of the RC can be improved by optimizing the shape of the silica fillers.

KEYWORDS

Resin composites, Filler shape, Nano-CT, Multi-scale analysis, Homogenization, Localization

1. INTRODUCTION

Posterior restorations have been increasingly performed with resin composites (RCs) to satisfy the aesthetic demands of patients [1]. However, the mechanical properties of RCs have some limitations compared with metal and ceramics [2]. RCs with universal shade, consisting of a resin matrix and glass filler, can be color matched to the tooth being restored, providing an aesthetic advantage [3, 4]. Newer compositions of these RC materials have been improved to withstand stress and wear [5]. RCs have been invented to closely mimic the aesthetics and function of natural tooth tissue, and their longevity in the oral environment depends largely on their fatigue or wear properties [6]. Identification of the mechanical properties of commercially-available RCs is difficult because of the variety of synthesis conditions. Clinical reviews have shown that the wear of RCs largely depend on the particle size and volume of filler or the resin matrix [7, 8]. However, other factors such as filler shape, filler distribution, coupling agent between filler and monomer, and filler/monomer composition ratio have not been considered.

Although the impact of the size and content of filler on the mechanical properties of RCs has been reported [9], the relationship between the mechanical properties and the RC micro-structure has not been clarified because the above other factors are influenced by one another and are not controllable as an independent parameter *in vitro*. Also, distinguishing nano-hybrids from micro-hybrids is difficult and their mechanical properties, such as flexural strength and modulus, tend to be similar [10]. In this study, the mechanical properties of RCs at the micro- and macro-scale were evaluated using *in silico* models with precise simulation of the micro-structure reconstructed based on high-resolution nano-CT images. Multiple factors, such as the amount of unpolymerized methacrylate monomer, the degree of hydrolysis at the interface between the resin matrix and filler [11], and temperature [12], are considered in an *in vitro* test. However, a method identifying the fracture initiation of materials has not been established and details of the 3D shape/structure of filler has not been investigated because of their small diameter.

In silico multi-scale analysis is a method for analyzing physical properties and their behavior among different size scales, which can be used to address issues in the characterization of heterogeneous substances by considering the material properties at the micro-scale [13]. *In silico* multi-scale analysis has been further enhanced to solve issues with the analysis of the mechanical properties of RCs [14] and the influence of the physical properties of an implant on cortical bone resorption [15].

The finite element analysis used in the *in silico* multi-scale analysis is a numerical approach to investigate the mechanical properties, and the maximum principal strain is used as a fracture criterion to predict the flexural strength of RCs [16, 17]. The utility of the maximum principal strain at the micro scale has not been reported because of the limited resolution of three-dimensional imaging technologies such as micro-CT to construct numerical model reflect to real morphology of fillers [9].

The aim of this study was to investigate the fracture criteria for predicting the fracture initiation of RCs at the micro scale and assess the influence of filler shapes on the flexural properties of RCs by combining nano-CT imaging and *in silico* multi-scale analysis.

2. MATERIALS AND METHODS

2.1 Sample preparation

An experimental RC containing irregular-shaped (IS) silica filler and 2,2-bis[4-(2-hydroxy-3-methacrylyloxypropoxy)-phenyl]-propane (Bis-GMA)/triethylene glycol dimethacrylate (TEGDMA) was prepared. The content (monomer/filler) of the experimental RC was 68.8/31.2 vol% (50.0/50.0 wt%). The monomer compositions were Bis-GMA (75 wt%), TEGDMA (25 wt%), benzoyl peroxide (BPO) (1.4 wt%), dl-camphorquinone (CQ) (0.7 wt%), and aldehyde compound with dibutylhydroxytoluene (2.17 wt%). IS silica glass (D99: 57.2 µm, D50: 5.7 µm) with a γ-methacryloxyproyl trimethoxysilane (MPS) surface modification was used as a filler.

2.2 *In vitro* three-point bending test

According to ISO 4049:2019, 25.0 × 2.0 × 2.0 mm bars were prepared (n = 8) for experimental RC containing IS silica filler. Each specimen was light cured for 10 s in nine spots on each side using a light-emitting diode curing light (Pencure 2000, Morita, Kyoto, Japan) with a maximal light density of 2,000 mW/cm^2. A three-point bending test was conducted and a mean fracture load was calculated using a universal testing machine (EZ-SX, Shimadzu, Kyoto, Japan) with a crosshead speed of 1.0 mm/min.

2.3 *In silico* models

The RC specimen containing IS silica filler was scanned by nano-CT (SkyScan1272, Bruker, Kontich, Belgium) at a resolution of 500 nm, and micro-scale models (100 × 100 × 100 μm) were randomly extracted from a scanned region (VG Studio MAX 2.0, Volume Graphics, Heidelberg, Germany) (Model 1 to Model 10) (Fig. 1). Another set of micro-scale models containing sphere-shaped (SS) filler with the same volume content as the experimental RC (31.2 vol%, D99: 57.2 μm, D50: 5.7 μm) were designed using CAD software (Solidworks Simulation 2011, Dassault Systèmes SolidWorks, Massachusetts, USA) (n = 10). A macro-scale model was designed simulating a three-point bending test consisting of three rods and a specimen using the same CAD software (Fig. 2). According to ISO 4049:2019, two parallel supporting rods were fixed with a span length of 20.0 mm and a third rod was centered between and parallel to the other rods. The three rods introduce three-point loading to the specimen (25.0×2.0×2.0 mm).

2.4 Homogenization analysis

The elastic moduli and Poisson's ratio for both the IS and SS silica filler micro-scale models were calculated by homogenization analysis using computer-aided engineering software (VOXELCON2015, Quint Corporation, Fuchu, JAPAN).

2.5 Failure criteria of the composite resins

The mean values of the elastic moduli and Poisson’s ratio were then applied to the macro-scale model. For simulation of the three-point bending test, a static load (the mean fracture load obtained from the *in vitro* test) was applied to the third rod in a downward direction at the center of the specimen. The maximum principal strain (MPStrain), maximum principal stress (MPStress), von Mises stress, and shear stresses in the YZ, ZX, and XY planes were evaluated as candidate failure criteria (n = 10). The standard deviation/mean for each failure criteria were calculated and compared.

After choosing the failure criterion that had the lowest standard deviation/mean, the flexural strengths of the RCs were predicted by finite element analysis using the macro-scale model and compared. The distribution of the failure criterion in the macro-scale model was assessed and the maximum values were calculated and compared.

2.6 Localization analysis

The distribution of the failure criteria in the micro-scale models was examined by localization analysis (VOXELCON2015, Quint, Fuchu, Japan). The localization analysis was conducted at the location in the macro-scale model where the maximum values of the failure criteria were observed.

2.7 Statistical analysis

The elastic modulus and Poisson's ratio values obtained from the homogenization analysis were statistically analyzed using two-way factorial ANOVA and a Bonferroni test (PASW Statistics 18, IBM, New York, USA). The flexural strength values obtained from the *in silico* three-point bending test were compared using a Student's *t*-test (PASW Statistics 18). P-values less than 0.05 were considered statistically significant.

3. RESULTS

3.1 Homogenization analysis

In the X, Y, and Z directions, significantly greater elastic modulus values were obtained for the RC models containing IS filler than with RC models containing SS filler ($p<0.05$) (Fig. 3a). In the Y and Z directions, significantly smaller Poisson's ratios were obtained for RC models containing IS filler than with RC models containing SS filler ($p<0.05$) (Fig. 3b).

3.2 *In silico* three-point bending test

The flexural strength of the RC models containing IS filler was significantly greater than RC models containing SS filler ($p<0.05$) (Fig. 4).

3.3 Failure criteria

The MPStrain was the lowest mean standard deviation of the failure criteria (Fig. 5) and was selected for the analysis.

3.4 Localization analysis

Figure 6 shows the MPStrain distribution in the YZ plane at the micro-scale for all models where the maximum MPStrain value was obtained in the macro-scale model. At the micro-scale, the maximum MPStrain value was localized within the resin matrix for all models (red arrow) (F: filler, M: resin matrix).

Figure 7 shows the micro-scale MPStrain distribution on the YZ plane for Model 1 where the maximum MPStrain value was obtained in the macro-scale (Upper row: SS filler, Lower row: IS filler). The red arrows indicate the location where the maximum MPStrain value was obtained in the micro-scale model. The red areas in the RC matrix under a static loading grew larger around the SS particles than the IS particles.

4. DISCUSSION

We established an *in silico* multi-scale analysis of RCs with different filler shapes (irregular/sphere). The multi-scale analysis clarifies the influence of the filler shape on the flexural properties of RCs.

Few systematic investigations have reported the effect of particle size and shape on RCs [18-22]. In the previous studies, the impact of the filler shape on the mechanical properties of RC were investigated using *in vitro* testing of various commercially-available products or experimental RCs in which variations, such as the filler size and the volume content of the filler, were not completely controlled. In this study, the designed *in silico* micro-scale models varied only in the geometric shape of the filler particles; under such conditions, the flexural strength can be predicted and compared. Therefore, any changes or trends in the mechanical performance can be attributed to the influence of the filler shape.

In terms of homogenization analysis, significantly smaller Poisson's ratios were obtained in the y and z directions of RC containing IS filler than RC containing SS filler. Although the IS and SS fillers were both homogeneously dispersed in the resin matrix, the results suggest the mechanical anisotropy of the RCs at the micro-scale. The relationship between the filler shape and elastic modulus was previously studied using an *in vitro* test, demonstrating that the RC with SS filler had lower elastic modulus than that with IS filler, irrespective of the filler size [23]. This is similar to the results in this study that used a constant mean value for the filler size. Poisson's ratio values of 0.3–0.4 have been reported for dental RCs [24-27]. In this study, the Poisson's ratio produced using the *in silico* method for both models ranged from 0.325 to 0.417.

The *in silico* three-point bending analysis successfully predicted the flexural strength using the mean load obtained from an *in vitro* test. The mean flexural strength of the RC containing IS filler prepared with the same mean filler size and volume content was reported as 114.8±10.5 MPa [16]. In this study, the flexural strength of RC model containing IS filler was 126.3±10.3 MPa, which is greater than that of RC model containing SS filler (104.9±6.5 MPa). This result suggests that the geometric shape of the filler does affect the flexural strength of the RCs under the same filler volume content. The greater mechanical performance of RC model containing IS filler can be attributed to the total surface area of the filler, where different shapes can have different surface areas [28]. IS filler has a 7.574 times larger surface area than SS filler. This surface area acts as an interface for stress transfer and the silane coupling rate increases with an increase in the total surface area of the filler [14, 29]. Composites that contain round filler particles have a high filler loading resulting in higher flexural properties than IS filler particles with intermediate filler loading [21]. For further study, the influence of the filler shape on the flexural strength should be evaluated in a simulation of a series of filler volume contents.

The MPS used in this study has been reported as a failure criterion to predict the flexural strength of RCs at the macro-scale [16]. All of the maximum MPS values obtained from the localization analysis were localized within the resin matrix in both micro-scale models of RC containing IS and SS silica filler because the models in this study were developed under the ideal condition that the filler and matrix were 100% bonded. The actual degree of conversion of the bonding agent between the filler and matrix is less than the ideal condition [30]. When the silane coupling ratios of the filler decrease, the maximum MPS value distribution moves to the matrix/filler interface where the silane coupling layer is located [31]. Within the limitation of the *in silico* static multi-scale analysis used in this study, the area where the maximum MPS value was observed in the SS silica filler micro-scale model increased around the filler compared with the RC containing IS filler, suggesting that a crack in a RC containing SS filler may more easily propagate than in a RC containing IS filler. The *in silico* non-linear dynamic FEA [17] will be useful to obtain more detailed interpretation for MPS distribution in micro-scale models of RC.

The *in silico* multi-scale analysis demonstrated that the filler shape has an effect on the flexural strength and MPS distribution of RCs under a constant filler volume content. Examination of the other physical properties, such as the fracture toughness and fracture strength influenced by filler shape, are also required. The fracture toughness and flexural strength of 14 commercially-available RCs classified by filler morphology have been reported; however, long-term *in vivo* trials to investigate the clinical performance are necessary [21]. A CAD/CAM RC model was successfully reconstructed from cryo-electron microscopy images and may have the potential to clarify the detailed mechanisms of how the filler shape influences the physical properties of RCs containing nano filler particles [30]. The combination of *in vitro* and *in silico* approaches established in this study will promote to develop new RCs and contribute to prolong the clinical performance/longevity of RCs.

5. CONCLUSIONS

An *in silico* multi-scale analysis demonstrated that RC models containing irregular-shaped filler had greater flexural strength than RC models loaded with sphere-shaped filler, suggesting that the mechanical strength of RCs can be improved by optimizing the shape of silica filler.

ACKNOWLEDGEMENTS

This research was supported by a Grant-in-Aid for Scientific Research (No. JP 19K10244) from the Japan Society for the Promotion of Science (JSPS). The authors are grateful to Kuraray Noritake Dental for kindly providing some of the materials used. We thank Ashleigh Cooper, PhD, from Edanz Group (https://en-author-services.edanzgroup.com/) for editing a draft of this manuscript.

FIGURE LEGENDS

Fig. 1 Nano-CT images and the reconstructed micro-scale model.

Fig. 2 Schematic illustration of the *in silico* multi-scale analysis consisting of homogenization and localization.

Fig. 3 a. Elastic modulus of resin composites (RCs) containing irregular-shaped filler and sphere-shaped filler. b. Poisson's ratio of RCs containing irregular-shaped filler and sphere-shaped filler.

Fig. 4 Flexural strength of the composite resins containing irregular-shaped filler and sphere-shaped filler.

Fig. 5 Percentage of the standard deviation/mean value for each failure criteria.

Fig. 6 Maximum principal strain (MPStrain) distribution of the YZ plane at the micro-scale for all models where the maximum MPStrain value was obtained in the macro-scale model (F: filler, M: resin matrix).

Fig. 7 Maximum principal strain (MPStrain) distribution in the YZ plane at the micro-scale for Model 1 at the location where the maximum MPStrain value was obtained in the macro-scale (Uppers: SS filler, Lowers: IS filler.). The red arrows indicate the locations where the maximum MPStrain value was obtained in the micro-scale model.

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