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Numerical evaluation of bulk material properties of dental composites using two-phase finite element models

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ABSTRACT

Objectives. The aim of this study was to numerically evaluate the effects of filler contents and resin properties on the material properties of dental composites utilizing realistic 3D micromechanical finite element models.
Methods. 3D micromechanical finite element models of dental composites containing irregular fillers with non-uniform sizes were created based on a large-scale, surrogate mixture fabricated from irregularly shaped stones and casting resin. The surrogate mixture was first scanned with a micro-CT scanner, and the images reassembled to produce a 3D finite element model. Different filler fractions were achieved by adjusting the matrix volume while keeping the fillers unchanged. Polymerization shrinkage, Young's modulus, Poisson's ratio and viscosity of the model composites were predicted using the finite element models, and their dependence on the filler fraction and material properties of the resin matrix were considered. Comparison of the numerical predictions with available experimental data and analytical models from the literature was performed.
Results. Increased filler fraction resulted in lower material shrinkage, higher Young's modulus, lower Poisson's ratio and higher viscosity in the composite. Predicted shrinkage and Young's modulus agreed well with the experimental data and analytical predictions. The McGee–McCullough model best fit the shrinkage and Young's modulus predicted by the finite element method. However, a new parameter, used as the exponent of the filler fraction, had to be introduced to the McGee–McCullough model to better match the predicted viscosity and Poisson's ratio with those from the finite element analysis.
Significance. Realistic micro-structural finite element models were successfully applied to study the effects of filler fraction and matrix properties on a wide range of mechanical properties of dental composites with irregular fillers. The results can be used to direct the design of such materials to achieve the desired mechanical properties.

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1. Introduction

Inorganic fillers provide dental resin composites with desirable properties such as high stiffness and hardness, and low coefficient of thermal expansion [1,2] and polymerization shrinkage [3]. However, not all material properties of dental composites can benefit from the addition of filler particles. For example, the viscosity of uncured composites increases dramatically with increasing filler fraction [4], leading to poor

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adaptation in the prepared cavities for dental restorations and a lower ability to relieve shrinkage stress. The development of shrinkage stress is considered to be a main cause for post-operative sensitivity and marginal leakage associated with composite restorations. A recent mathematical analysis showed the relative importance of the different material properties of dental composites in the build-up of shrinkage stress, with polymerization shrinkage being the most important, followed by Young's modulus and viscosity [5]. Thus, despite reduced polymerization shrinkage, increased filler content in dental composites may actually increase the shrinkage stress because of increased Young's modulus and viscosity [6–8]. Therefore, understanding the effect of filler particles on the above material parameters of dental composites is essential in selecting an appropriate filler volume to minimize shrinkage stress in composite restorations.

The dependence of the material properties of dental composites on the filler volume may be studied directly by testing a series of these materials with various filler fractions [2,9–12]. However, available commercial materials have different compositions, the properties of which are not always known. Even if composites with known compositions are employed, it will still be difficult to control the degree of conversion in the resin matrix among samples with different filler fractions because of irradiation shielding by the fillers. The resulting uncertainties in the matrix condition will prevent the correct relationships between the filler fraction and bulk material properties of the composite from being determined. Furthermore, preparing the samples and performing the various tests to determine the material properties are very time-consuming.

Alternatively, analytical or numerical methods can be used to predict the material properties free of experimental errors and uncertainties. For example, Lingois and Berglund [10] compared predictions from existing micromechanical models with data for experimental dental composites of different filler volume fractions. They observed that, for shrinkage, predictions given by the Rosen–Hashin model agreed well with experiments. For the elastic constants, semi-empirical models, such as Halpin–Tsai and that of Chantler et al. [13], provided good predictions. However, most analytical models require simplifications with regard to the filler particle shape, size and spatial distribution, which often lead to disagreement between predictions and experimental data. For example, at very high filler volumes, some of the analytical models under-predict the elastic constants, possibly because of contact interactions between the filler particles that are not accounted for.

These shortcomings of analytical models can be overcome by using geometrically realistic micromechanical models such as those based on the finite element method (FEM). A few applications of FEM to determine the elastic properties of composites with different volumetric fractions of randomly distributed fillers (short fibers and spherical particles) have been reported in the literature [14–16]. For example, the finite element model used by Sun et al. [15] contains spherical fillers of a uniform size randomly distributed within a BIS-GMA/TEGDMA resin matrix.

Of course, most dental composites do not contain fillers of a regular shape and size. The aim of this study is therefore to use FEM to explore the effects of the filler volume, as well as the material properties of the resin matrix, on the bulk properties of composites that contain randomly distributed fillers of irregular shapes and sizes. Geometrically realistic, 3D micromechanical FE models based on micro-CT images of a macro composite with a similar 2-phase construct were employed to predict the mechanical properties of such dental composites. The numerical results were compared with predictions from several analytical models.

2. Materials and methods

2.1. *Modeling composites reinforced with irregular fillers*

The models of composites reinforced with irregular filler particles used in this study were derived from a much larger-scale surrogate mixture of irregularly shaped stones and a casting polyester resin which consists of MetPrep Kleer-Set Type FF resin and MetPrep Kleer-Set Hardener (Metprep Ltd., Coventry, UK). The stones were crushed decorative gravels that have shapes similar to the irregular fillers in some dental composites [1]. At first, 20 cm^3 of the casting resin was mixed with a hardener in a paper cup according to the manufacturer's instructions. The stones were then blended into the resin to produce a mixture of $\sim$40 cm^3 in volume. The mixture was stirred continuously until the resin began to set to make sure the stones were dispersed evenly. About 1 h, when the resin had hardened, the block was removed from the cup and cut into a rectangular shape using a diamond cutter, as shown in Fig. 1(a). In total, there were 167 stones in the block, giving a volumetric filler fraction of $\sim$50%. The maximum, minimum and mean size of the stones in the longest dimension were 6.0 mm, 1.6 mm and 4.2 mm, respectively; and the size of the block after machining was 13 mm × 13 mm × 15 mm.

The composite block was then scanned with a micro-CT scanner (SkyScan 1072, SKYSCAN, Belgium); see Fig. 1(b). Because of the difference in density, and hence grayscale value, the stones could easily be separated from the resin during the reconstruction of the 3D model (Fig. 1(c)) using commercial software (ScanIP, Simpleware Ltd., Exeter, UK). However, partition by hand was still necessary in some areas where the boundaries of the stones were not clear. Thereafter, the 3D spatial model was converted into a 3D finite element model (Fig. 1(d)) using another commercial software (ScanFE, Simpleware Ltd., Exeter, UK). Smoothing was performed on the boundaries of the stones during the construction of finite element model in order to avoid highly distorted elements. The fillers in the model therefore had more rounded edges than the actual stones; see Fig. 1(d).

2.2. *FE models of dental composites with different filler fractions*

The filler fraction of the first FE model created was approximately 50%. To obtain other filler fractions, preliminary finite element analysis (FEA) was carried out by either expanding or compressing the matrices while keeping the fillers undeformed. Thus, the fillers were defined as rigid bodies while