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# Application of close-packed structures in dental resin composites

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## ABSTRACT

**Objective.** The inorganic filler particles in dental resin composites serve to improve their mechanical properties and reduce polymerization shrinkage during their use. Efforts have been made in academia and industry to increase the filler particle content, but, few studies examine the theoretical basis for the maximum particle loading.

**Methods.** This work evaluates the packing of spherical particles in a close-packed state for highly loaded composites.

**Results.** Calculations show that for low dispersity particles, the maximum amount of particles is 74.05 vol%, regardless of the particle size. This can be further improved by using a mix of large and small particles or by the use of non-spherical particles. For representative spherical particles with a diameter of 1000 nm, two types of secondary particles with respective sizes of 414 nm ( $d_I$ ) and 225 nm ( $d_{II}$ ) are selected. The results show that after embedding secondary particles I & II into primary spherical particles, the packing factor is increased to 81.19% for the close-packed structures, which shows an improvement of 9.64%, compared to the 74.05% obtained only with primary spherical particles. This packing factor is also higher than either structure with the embedded secondary particles I or II.

**Significance.** Examples of these mixtures with different spherical particle sizes are shown as a theoretical estimation, serving as a guideline for the design and formulation of new dental resin composites with better properties and improved performance.

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## 1. Introduction

Dental resin composites are mainly comprised of an organic matrix mixed with silanized inorganic fillers, and can be cured to form a crosslinked composite polymer network with the blue light irradiation [1]. While the resin phase contributes to the toughness of the composites related to the mechanical strength in flexure or tension, their mechanical properties in regard to their modulus and hardness, are dominated by the filler particles for the similar resin matrices [2,3]. Recent research efforts have been aimed at optimizing filler formulations including filler loading, size, and morphology, with the

aim of improving mechanical properties and reducing polymerization shrinkage during their clinical use [4–6]. These results have shown that higher loading fractions in resin composite have led to materials with superior mechanical properties. For filler formulations with larger particles, the addition of smaller particles, either spherical or irregular, could improve fracture toughness, shrinkage strain, and wear behavior of the resulting composites [7–9]. However, in previous reports, authors show series of filler formulations where the reason for choosing them is seldom mentioned [7–9]. Few studies examine the theoretical basis for the maximum particle loading in a resin composite [10]. Therefore, it is desirable and useful to develop a model for the refinement of filler formulations without trial and error.

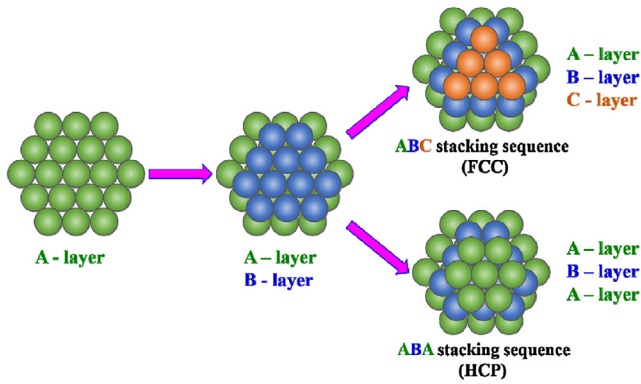
This model in this paper is based on the theory of close-packed structures of spherical particles of identical sizes for

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**Fig. 1 – Stacking sequences of face-centered cubic (FCC) arrangement and hexagonal close-packed (HCP) arrangement.**

the refinement of filler formulations with the aim to estimate the maximum allowed loading of the particles.

## 2. Methods

### 2.1. Particle packing theory

The following particle optimizations are based on previous work on systems modeling the packing of identical spheres. Most of the previous work was first explored in the context of crystal structures, which showed that atoms tend to arrange in certain patterns, or lattice arrangements, the most efficient of which are known as close-packed structures. These are modeled with a unit cell that is repeated in all directions for the purpose of packing, though in some cases boundary conditions may need to be defined. The two close-packed arrangements with the highest packing efficiency are called face centered cubic (FCC) and hexagonal close packed (HCP) [11,12]. Their stacking arrangements and unit cells are shown in Figs. 1 and 2, respectively. While most of the work on packing structures applies to atoms or nanoparticles (on the order of 10 nm) [13], the lower mobility of larger particles such as those used for dental composites may not allow effective FCC and HCP arrangements, resulting in random packing orders. For the purpose of modeling voids in the structures, FCC and HCP arrangements are used since they are the most appropriate.

In the literature, the particle loading for dental resin composites is most often quoted as the mass fraction of the particles with respect to the total mass of the final product. Since the density of the filler in dental materials is much higher than that of the resin, typically on the order of 2.2 g/cm<sup>3</sup> for silica [14] vs 1.161 g/cm<sup>3</sup> for bisphenol A glycerolate dimethacrylate (Bis-GMA) [15], the weight fraction is higher than the volume fraction for the filler loading. In the context of sphere packing, the packing factor instead refers to the volume fraction occupied by particles in the unit cell, which is defined as [11]:

$$\text{Packing factor} = V_s/V_c \quad (1)$$

where  $V_s$  is the volume of all the spheres in a unit cell (assuming the hard sphere model), and  $V_c$  is the total volume of a unit cell.

#### 2.1.1. FCC

The FCC arrangement is based on a cubic unit cell with particles located at each of the corners and at the centers of each the square face [11], as shown in Fig. 2(a–c). As previously studied, a total of four whole particles can be assigned to a FCC unit cell (Table 1), therefore, their occupied volume can be defined as  $V_s = (4 \times \frac{4\pi}{3}R^3)$ , where  $R$  is the radius of particles. The total volume of the unit cell is computed as  $V_c = a^3$ , where  $a$  is the length of the unit cell (Fig. 3a). Following simplification, the unit cell size  $a$  and the particle radius  $R$  cancel out, resulting in a size-independent packing factor of  $\frac{\sqrt{2}\pi}{6}$ , or approximately 74.05% [16–18].

#### 2.1.2. HCP

Similarly, the HCP arrangement is based on a stacked hexagonal arrangement of particles, with a particle at each vertex of the hexagon, in addition to one in the center. Two of these are stacked with an intervening layer that provides three additional particles (Fig. 2d–f) [11]. Still using particles of radius  $R$ , and seeing a total of six whole particles for a unit cell (Table 1), thus  $V_s = (6 \times \frac{4\pi}{3}R^3)$ , and  $V_c = (6 \times \frac{1}{2} \times \frac{\sqrt{3}}{2}a^2 \times c)$ , where  $a$  and  $c$  represent the short and long unit cell dimensions, and calculations yield the same value as FCC, with an identical packing factor of 74.05% (Fig. 3b) [17,18].

The above results indicate that, regardless of particle size, even in the most efficient packing arrangements, identical spheres cannot completely fill the space. There are necessarily voids between particles; these voids have complex shapes, however for filler loading purposes, we are mostly interested in the largest spherical particles which can fit into these voids. In the close-packed structures discussed above, two types of voids are formed, tetrahedral voids and octahedral voids, which are identical in both FCC and HCP as the voids exist between two layers of atoms (Fig. 4) [19]. However, the number of voids in these close-packed structures is different, which will be addressed further. Previous work has also shown that the maximum diameter of a tetrahedral void ( $d_4$ ) is 0.45  $R$ , whereas that of an octahedral void ( $d_8$ ) is 0.828  $R$ , where  $R$  is the radius of the packed spheres, as derived from the close packing sphere models [20].

#### 2.1.3. RCP

While FCC and HCP are the most efficient packing arrangements, many more exist, such as simple cubic and body centered cubic (BCC) [11,21]; their detailed information including coordination numbers, number of particles per unit cell, and packing efficiency are collected in Table 1 [21,22]. However, these fully ordered models assume frictionless interactions between the particles, such that diffusion is very fast on the scale of the particles. In order to model systems in which friction is a non-negligible force and particle diffusion is slow, random packing is considered. When a set of spheres is put into a packed state, a random packed arrangement will generally result, and there exists a range of random-packing packing factors, with an upper limit of 64% and a lower limit of 55%

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