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Correlation of resin viscosity and monomer conversion to filler particle size in dental composites

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ABSTRACT

Objective. The viscosity of dental resin composites is important in their formulation and clinical use; it depends on the filler particle size and loading. We intend to study the viscosity and conversion of composites made of low dispersity spherical silica fillers.

Methods. Experimental dental resin composites were formulated using low dispersity spherical silica particles of graded sizes (75, 150, 500, 350, 500, 1000 nm) at several loading levels with resins based on Bis-GMA and UDMA. Their rheological properties and double bond conversion were measured with a rheometer and differential scanning calorimeter, respectively.

Results. The complex viscosity of the unpolymerized pastes can be fit to an extended Krieger–Dougherty equation that includes an adjustment factor to account for filler particle surface area. This relationship is also extended to estimate the degree of conversion, where the calculated or experimental viscosity is used to predict the resulting conversion.

Significance. The enhanced understanding of the relationship of filler size, composite viscosity, and monomer conversion will allow improved accuracy in the prediction of the properties of dental resin composite formulations to obtain ideal viscosity for their clinical use and a high degree of conversion.

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1. Introduction

Dental resin composites have experienced a significant rise in usage since their advent in the 1960s. These composites are made from a photopolymerizable resin matrix (including initiator, co-initiator and inhibitors) mixed with surface-modified inorganic fillers. Most of the organic resin monomers in these products are based on the commercially available bisphenol A glycerolate dimethacrylate (BisGMA), diurethane

dimethacrylate (UDMA), and triethylene glycol dimethacrylate (TEGDMA). The inorganic fillers used in commercial dental restoratives are generally silicates, most commonly silica or alkaline glasses [1]. Many companies produce composites, and a large body of literature exists studying their mechanical properties and conversion values [2,3]. The rheological properties and ease of polymerization are significant factors affecting the clinical use of these materials, but few studies have systematically examined the effect of filler particles on composite viscosity, and its relationship to the dental monomer conversion.

The viscosity of these materials is important for commercial formulations since it determines the ease of use of resin

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composites [4]. The viscosity is also well known to affect the degree of polymerization and reaction kinetics in free radical polymerizations due to the reduced chain mobility and lower termination rate at higher viscosities [5]. While the effect of resin monomer composition on viscosity has been explored extensively [6,7], viscosity increases due to the addition of filler particles is not as well characterized, particularly for the high loading levels where particle-particle interactions become more important. To the best of our knowledge, no previous work has explored narrow dispersity fillers of different sizes with the same morphology to clearly define this thickening effect.

The degree of conversion in dental composites is related to their mechanical properties such as diametral tensile strength and flexural strength [8,9]. Most of the work so far has examined the conversion of resin matrix alone [7,10], and the effect of different monomer compositions on the conversion and viscosity. Turssi et al. [11] found that filler size and morphology did not affect conversion except when the size corresponded to the photopolymerization wavelength, where a lower conversion was observed. Otherwise, the filler loading was shown to affect the shrinkage and mechanical properties after polymerization [12,13]. Others examined the degree of conversion of different commercial formulations for which the exact composition was not known [14,15], providing limited assistance in establishing fundamental rules for composite design.

The detailed formulations of commercial dental composites remain proprietary to the manufacturers, therefore the full effect of filler particles on composite properties before and after polymerization has not yet been established under controlled conditions. There remains a lack of fundamental understanding of the factors influencing the mechanical, chemical, and optical properties of the composites. We intend to establish a relationship between the size of the filler particles and the viscosity of dental composites, and in turn, the effect on the monomer conversion during photopolymerization.

2. Experimental

2.1. Materials

Tetraethyl orthosilicate (TEOS) was purchased from Alfa Aesar (Ward Hill, MA, USA). Anhydrous ethanol and ammonium hydroxide (35%) were purchased from Fisher Scientific (Waltham, MA, USA), 3-methacryloyltrimethoxypropylsilane (3-MPS), BisGMA, UDMA, and TEGDMA were purchased from VWR (Mont-Royal, QC, Canada); camphorquinone (CQ) and ethyldimethylaminobenzoate (EDMAB) were purchased from Sigma-Aldrich (Oakville, ON, Canada). All reagents were used without further purification. Room temperature is defined as 23 °C in this work.

2.2. Methods

2.2.1. Synthesis of monodisperse silica particles and characterisation

The silica particles with sizes of 75, 150, 350, 500, 1000 nm were synthesized by using the Stöber method [16] as described pre-

viously [17]. The sizes indicated in figures and tables are not the nominal sizes but refer to the particle diameters measured by laser diffraction (Horiba LA960, Japan), and scanning electron microscopy (JEOL FE-SEM, JSM-7400F, Japan) for the specific batches of particles that were used.

Thermogravimetric analysis (TGA 2950 from TA Instruments, DE, USA) was used to measure the extent of silane surface modification. The temperature was increased to 120 °C for 10 min to eliminate adsorbed water, and then a 20 °C min⁻¹ ramp to 800 °C was then used to obtain the extent of silane modification.

The surface area of the particles was calculated from first principles using sphere area and volume equations, and confirmed with previous BET measurements [18], adjusted for the particle size. The equation is derived from the ratio of surface area in relation to volume and density:

$$S = \frac{A_{\text{sphere}} \Phi}{V_{\text{sphere}} \rho} = \frac{\pi d^2 \Phi}{\frac{1}{6} \pi d^3 \rho} = \frac{6\Phi}{d\rho} \quad (1)$$

where S is the specific surface area (m²g⁻¹) of the fillers, A_{sphere} and V_{sphere} are the surface area and volume of a sphere, Φ is the mass fraction of the fillers, ρ is the density of fused silica (2.2×10^6 g m⁻³ [19]), and d is the particle diameter (m).

2.2.2. Resin and composite preparation

70:30 weight ratio BisGMA-TEGDMA (7B3T) and UDMA-TEGDMA (7U3T) resins were each blended by first dissolving 0.5 wt% CQ and 0.5 wt% EDMAB in TEGDMA (as a fraction of total resin weight) at room temperature, then adding the main monomer (either BisGMA or UDMA), mixing manually and then with a magnetic stirrer at 35 °C (approx. 30 min) to obtain a homogeneous mixture. Please note the first letters B, T and U are used represent the three monomers in the blends with the weight ratio indicated in numbers. The resulting resin was then mixed with the indicated amount of filler particles (30, 50, 60, or 70 wt%) by spatula, and then homogenized using a three-roll mill (Exact 50i TRM, Norderstedt, Germany). Only the composites used for conversion measurements contained initiator (CQ) and co-initiator (EDMAB).

2.2.3. Rheological studies

The rheology measurements were performed using an AR-2000 shear rheometer (TA Instruments) with 20 mm parallel steel disk and plate geometry with a gap size of 200 μm for 60 wt% loading and 1000 μm for 70 wt% loading. The composite pastes were put in place and trimmed using a spatula. The measurements were performed with a dynamic oscillatory shear test, at a constant frequency of 1 Hz, and time series at constant stress (10 Pa) where thixotropic behaviour was observed. Each measurement was repeated three times, with a recovery period between each run to allow full recovery. The complex viscosity defines the internal friction of a material under oscillatory shearing stresses; the values shown are the magnitude of the complex viscosity vector ($|\eta^*|$), hereby referred to simply as 'complex viscosity':

$$\eta' = \frac{\sigma}{\dot{\gamma}} \cos \theta \quad (2)$$

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