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Synthesis and study of properties of dental resin composites with different nanosilica particles size

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ABSTRACT

Objectives. The aim of this work was the synthesis of light-cured resin nanocomposites using nanosilica particles with different particle size and the study of some physical–mechanical properties of the composites.

Methods. Various types of silica nanoparticles (Aerosil) with average particle size of 40, 20, 16, 14, and 7 nm, used as filler were silanized with the silane 3-methacryloxypropyltrimethoxysilane (MPS). The total amount of silane used was kept constant at 10 wt% relative to the filler weight to ensure the complete silanization of nanoparticles. The silanizated silica nanoparticles were identified by FT-IR spectroscopy and thermogravimetric analysis (TGA). Then the silanized nanoparticles (55 wt%) were mixed with a photoactivated Bis-GMA/TEGDMA (50/50 wt/wt) matrix. Degree of conversion of composites was determined by FT-IR analysis. The static flexural strength and flexural modulus were measured using a three-point bending set up. The dynamic thermomechanical properties were determined by dynamic mechanical analyzer (DMA). Sorption, solubility and volumetric change were determined after storage of composites in water or ethanol/water solution 75 vol% for 30 days. The TGA for composites was performed in nitrogen atmosphere from 30 to 700 °C.

Results. As the average silica particle size decreases, the percentage amount of MPS attached on the silica surface increases. However, the number of MPS molecules attached on the silica surface area of 1 nm^2 is independent of filler particle size. As the average filler particles size decreases a progressive increase in the degree of conversion of composites and an increase in the amount of sorbed water is observed.

Significance. The prepared composites containing different amount of silica filler, with different particle size, but with the same amount of silanized silica and organic matrix showed similar flexural strength and flexural modulus, except composite with the lowest filler particle size, which showed lower flexural modulus.

1. Introduction

The introduction of resin-based composite technology to restorative dentistry was one of the most significant contributions to dentistry in the last century. Nowadays, esthetic tooth-shaded dental restorations are well-accepted and becoming more and more popular over metallic dental amalgams. Dental resin composites are consisted of a polymeric matrix admixed with silane reinforcing inorganic filler. Silane

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provides a crucial link between the matrix and the filler that can have a significant effect on the overall-performance of composite [1]. The organic matrix is based on methacrylate chemistry, especially cross-linking dimethacrylates [2–7], while the fillers have different type (silica, ceramic, etc.), size, shape and morphology [1,6,8]. Because of the major influence of the fillers on the physical properties, the classification of dental filling composites is based on the type and the particle size of fillers [1,5,6,8,9]. In general, two types of composites are available on the market, the microfill and the hybrid composites. Microfill composites were formulated having fillers with an average particle size in the range of 0.01–0.05 μm and were launched in the market to overcome the problems of poor esthetic properties. However, the mechanical properties are considered low for application in regions of high occlusal forces. Hybrids, on the other hand, offer intermediate esthetic properties, but excellent mechanical properties by the incorporation of fillers with different average particle sizes (15–20 μm and 0.01–0.05 μm) [8].

In dentistry, posterior restorations (class I or II) require composites that show higher mechanical properties, while anterior restorations (class IV–V) need composites that have superior esthetics. The resin composite that meets all the requirements of both posterior and anterior restorations has not emerged yet. Therefore, the great interest in resin composite's research is nanotechnology. Dental nanocomposites or nanofilled dental composites are claimed to combine the good mechanical strength of the hybrids [5,10,11] and the superior polish of the microfills [12]. Because of the reduced dimension of the particles (0.1–100 nm) and their wide size distribution, an increased filler load can be achieved technically resulting in reducing the polymerization shrinkage [10,14] and increasing the mechanical properties, such as tensile strength, compressive strength and resistance to fracture. On the other hand, the small size of filler particles can improve the optical properties of resin composites, because their diameter is a fraction of the wavelength of visible light (0.4–0.8 μm), resulting in the human's eye inability to detect the particles [11]. Furthermore, the wear rate is diminished and the gloss retention is better [11,13,15]. As a consequence, manufacturers now recommend the use of nanocomposites for both anterior and posterior restorations. Nowadays, nanocomposites are available as nanohybrid types containing milled glass fillers and discrete nanoparticles (40–50 nm); and as nanofill types containing both, nanosized filler particles called nanomers, and agglomerations of these particles described as "nanoclusters" [11]. The nanoclusters provide a distinct reinforcing mechanism compared with the microfill or nanohybrid systems resulting in significant improvements to the strength and clinical reliability [16].

This study was designed to examine the effect of nanosilica particle size, which is silane reinforced, on some selected physico-mechanical properties of light-cured composites based on Bis-GMA/TEGDMA (50/50 wt/wt) matrix. Silica nanoparticles, used as filler, have a controlled size from 7 to 40 nm. Their surface was treated with the silane coupling agent 3-methacryloxypropyltrimethoxysilane (MPS).

2. Materials and methods

2.1. *Materials*

Propylamine, 99+% (Lot no 0.4419MS), cyclohexane, 99+% (Lot no S22455-155), and MPS, 98% (Lot no. 09426EC-265) used in the silanization of nanoparticles of silica were received from Sigma–Aldrich GmbH (Deisenhofen, Germany). The monomers used, i.e. 2,2-bis[4-(2-hydroxymethacryloxypropoxy) phenyl]propane (Bis-GMA) (Lot no 07210BB) and triethyleneglycol dimethacrylate (TEGDMA) (Lot no 09004BC-275) were provided also from Sigma–Aldrich. The photoinitiator systems were camphorquinone, 97% (CQ) (Lot no S12442-053) and ethyl 4-dimethylaminobenzoate, 99+% (4EDMAB) (Lot no: 90909001) from Sigma–Aldrich. The fillers (Aerosil OX50, Aerosil 90, Aerosil 130, Aerosil 150, and Aerosil 300) used were from Degussa AG (Hanau, Germany). They are fumed amorphous silica with different average specific surface area (determined by the BET gas adsorption technique; BET comes from initial letters of authors' names who proposed it, i.e. Brunauer, Emmett, and Teller) and average particle diameter. Their specifications are listed in Table 1. All the materials used in this study were utilized as received, without further purification.

2.2. *Silanization of silica nanoparticles*

The Aerosil OX50, Aerosil 90, Aerosil 130, Aerosil 150 and Aerosil 300 silica nanoparticles were silanized with MPS, following the method of Chen and Brauer [17]. In all cases, the amount of organosilane was kept constant at 10% wt/wt relative to silica, a ratio that was more than enough to completely cover the surface of the silicas and to provide a durable interphase [18]. The amount of silane in grams (g) to obtain a minimum uniform coverage of the filler particles (X) is given by the following equation [18,19]:

$$X = \left(\frac{A}{\omega}\right) f$$

In which A is the surface area of the filler (50, 90, 130, 150, and 300 m^2/g), ω is the surface coverage per gram of silane MPS ($\omega = 2525$ m^2/g) [20] and f is the amount of silica (g).

The silica (5.0 ± 0.05 g), the silane (0.50 ± 0.01 g), the solvent (100 ml cyclohexane) and n-propylamine (0.1 ± 0.01 g) were stirred at room temperature for 30 min and then at 60 ± 5 °C for additional 30 min at atmospheric pressure. The mixture was then placed in a rotary evaporator at 60 °C for removing the solvent and volatile by-products. The powder was then heated at 95 ± 5 °C for 1 h in a rotary evaporator and finally was dried at 80 °C in a vacuum oven for 20 h.

The silanized silica nanoparticles were identified by FTIR spectroscopy (FTIR Spectrum One, Perkin Elmer, resolution 4 cm^{-1}, 32 scans, 4000–1350 cm^{-1}). Spectroscopic grade potassium bromide (KBr, Sigma–Aldrich, Cas no 7758-02-3) and silica powder were pressed together into a pellet using a KBr palletizer. Thermogravimetric analysis of the silanized silica nanoparticles was performed using a Pyris 1 TGA (Perkin Elmer) thermal analyzer utilizing about 5 mg of each sample.