



## Effect of Silanized Silicon Dioxide and Titanium Dioxide Nanoparticles Loading on the Compressive Strength of Prepared Composite Core Material

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### ABSTRACT

**Purpose:** the aim of this study was to assess the effect of silanized silicon dioxide and titanium dioxide nanoparticles loading on the compressive strength of experimentally prepared resin composite core material. **Materials and Methods:** Resin matrix was prepared by adding 70 wt% Bis-GMA, 30 wt% TEGDMA, 0.5 wt% CQ and 0.5 wt% EDAB. Sintering of the silica nanoparticles was performed and then the silanized sintered silica fillers together with titanium dioxide nanoparticles and silica nanoparticles were mixed with the prepared resin matrix. Specimens of the experimental and control resin composite were prepared in split Teflon moulds (4x6mm) for compressive strength testing using universal testing machine. **Results:** There was no statistically significant difference between results of the experimental and control resin composites. **Conclusion:** Addition of up to 85 wt% of nanofillers into traditional resin matrix produced a resin composite with comparable compressive strength to that of the commercial composite products commonly used as core material.

### INTRODUCTION

The ultimate target of dental restorations is to restore the biological functions and successful esthetics. Resin composites simplified this target by their satisfactory performance guided by their conservation to tooth tissues and good esthetic properties. They are also used as core materials with the challenge to function in high stress bearing areas.

### KEYWORDS

*Bis-GMA, TEGDMA, EDAB*

- Paper extracted from PhD Thesis titled "Effect of Silanized Silicon dioxide and Titanium Dioxide Nanoparticles Loading on Some Properties of Experimentally Prepared Composite Core Material".
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Extensive studies have been carried out to improve the performance of dental resin composites by modifying their chemical compositions. Among these improvements was the addition of different types of nanofillers. Nanomaterials continue to be a challenge in different medical fields as well as in dentistry, supported by their specific criteria of excellent handling properties, mechanics, biocompatibility and esthetics. Nanofillers are categorized either as isolated particles, with dimensions of nearly 1 to 100nm, or fused aggregates<sup>(1-4)</sup>.

Titanium dioxide nanoparticles have been investigated and shown effective multifunctional properties. It was found that, as their size decrease, especially below 50nm, they show special photo-induced activities originating from the semiconductor band gap. While, the energy becomes higher than the band gap, photons produce electron pairs and energy is released that could be activated by appropriate light source. Furthermore, TiO<sub>2</sub> nanoparticles also had excellent mechanical properties with high elastic modulus 230 GPa<sup>(5,6)</sup>.

While, silica nanoparticles have been used in the

medical field; in biosensors, drug delivery systems and in the treatment of tumors. The effect of addition of nanosilica into resin composites revealed reduced curing time, improved optical properties and perishability, in addition to, improved mechanical properties; tensile strength, shear strengths and microhardness<sup>(7,8)</sup>.

Several trials and research works were devoted to improve the properties and function of resin composite restorations, however very little researches were done to prepare resin composite core material totally filled with silica and titanium dioxide nanoparticles. Therefore, the present study was performed to prepare an experimental resin composite core material loaded with silanized silicon dioxide and titanium dioxide nanoparticles and assess its compressive strength.

## MATERIALS AND METHODS

Commercial resin composite material and chemicals used in the current study are listed in tables (1) and (2), respectively.

**Table (1):** Commercial name, manufacturer and batch number of the commercial material used in the study.

No.	Material / Commercial Name	Manufacturer	Composition	Batch Number
1	Nanohybrid resin composite/ Nexcomp	META BIOMED, Korea	Resin: Bis-GMA, Bis-EMA,UDMA,TEGDMA- Fillers: 0.04-0.7 $\mu$ m barium aluminum boro silicate	NXC1608101

**Table (2):** Chemical name, presentation, manufacturer and batch number of chemicals used in preparation of the experimental resin composite.

No.	Chemical name	Presentation	Manufacturer	Batch Number
1	Bisphenol A glycerolate dimethacrylate (Bis-GMA)	Viscous liquid	Sigma Aldrich, USA	MKBS3691V
2	Triethylene glycol dimethacrylate (TEGDMA)	Liquid	Sigma Aldrich, USA	STBF2613V
3	Trimethoxy silane	Liquid	Sigma Aldrich, USA	MKBH2196V
4	Silicone dioxide nano powder (spherical) (SiO <sub>2</sub> )	Powder (size: 5-15 nm)	Sigma Aldrich, USA	637246
5	Titanium dioxide nanopowder (TiO <sub>2</sub> )	Powder 32nm	Alfa Aesar	P10B008
6	Ethyl 4-(dimethylamino) benzoate(C <sub>11</sub> H <sub>15</sub> NO <sub>2</sub> )	Powder	Sigma Aldrich, USA	E24905
7	Camphorquinone	Powder	Alpha Aesar, USA	A14967

**Grouping of the specimens:**

Ten specimens were prepared for the laboratory study. The specimens were divided into two groups, the control group; (Nexcomp nanohybrid core resin composite (n=5) and the experimental group (the experimental composite resin core material for the assessment of compressive strength).

**Preparation of the experimental resin composite:*****Preparation of the resin matrix:***

The monomer was prepared by mixing 70 wt% Bisphenol-A glycerolate dimethacrylate (Bis-GMA), and 30 wt% triethylene glycol dimethacrylate monomers (TEGDMA). For the preparation of the photo-initiator system, 0.5wt% camphor quinone and 0.5wt% ethyl 4-dimethyl-amino benzoate (EDAB) were weighed, then added gradually to the prepared matrix and stirred for 1 hour<sup>(9)</sup>.

***Sintering of silica nanoparticles:***

Sintering of the silica nanopowder was performed by heating the silica nanopowder in an electric oven (Carbolite CWf 12/13, England) at 1300°C for 15 minutes at heating rate of 20 °C/ min. The sintered particles were ball milled then sieved with mesh no. 400<sup>(9)</sup>.

**Silanization and incorporation of the nanoparticles:**

Silane coupling agent was prepared by proportioning 70 wt % of ethanol in glass beaker, then few drops of acetic acid were then added gradually into it to decrease the pH to (3–4). Finally 3 wt % of trimethoxysilane was added into the solution. Stirring was done using a magnetic stirrer for one hour. Sintered silicone dioxide nanoparticles (80 wt%), titanium dioxide nanopowder (1wt%) , and silicone dioxide nanoparticles( 4 wt %) were silanized for 2 hours then centrifuged (centrifuge model 800, China) for 30 minutes. Finally, the precipitate was dried for 1 hour in hot oven at 110° C. The silanized fillers were incrementally added to the experimentally prepared resin matrix with a plastic spatula to form a homogenous composite paste<sup>(9,10)</sup>.

**Characterization of the sintered silica:**

Sintered silica was observed by means of a transmission electron microscope (JEM-1400, JEOL Ltd., Japan) before and after silanization.

**Compressive strength measurement:**

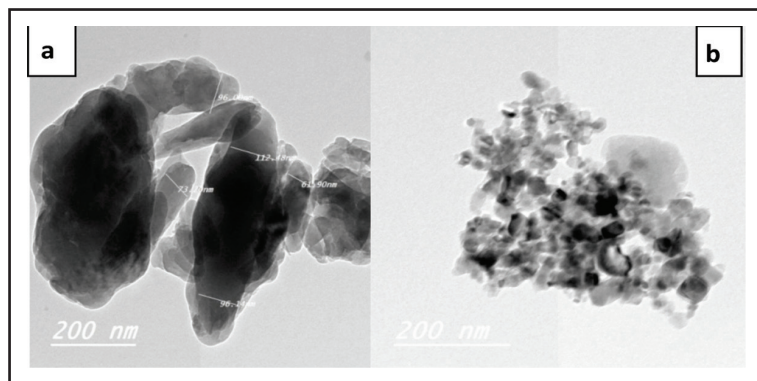
Five specimens for each resin composite type was prepared for compressive strength (CS) testing using cylindrical Teflon moulds 4mm in diameter and 6 mm in height according to ADA specification no. 27<sup>(11)</sup>. Each specimen was incrementally packed in the mould and cured on a glass slab placed over celluloid strip. Another celluloid paper was pressed at the top against another glass slab to extrude excess material. Curing was done using light-emitting diode (LED) curing unit (RTA, MINIS, Guilin Woodpecker Medical Instrument Co. Ltd., China): 1000-1200 mW/cm<sup>2</sup> for 40 seconds. Excess material was removed using Soflex discs. Compressive strength testing was done after immersion of the specimens in distilled water for 24 hours using universal testing machine (AGX- PLUS, SHIMADZU, 5KN) with 50 N load cell and crosshead speed 0.5 cm/min<sup>(12)</sup>.

**Statistical Analysis**

The mean and standard deviation values were calculated for each group in each test. Data were explored for normality using Kolmogorov-Smirnov and Shapiro-Wilk tests, data showed parametric (normal) distribution. Independent sample t-test was used to compare between two groups in non-related samples. The significance level was set at  $P \leq 0.05$ . Statistical analysis was performed with IBM® SPSS® Statistics Version 20 for Windows.

**RESULTS**

Results of TEM images, as shown in (Figure 1), showed distinguished homogenous distribution of the sintered silica clusters. The clusters appear in more uniform pattern after the silanization process.

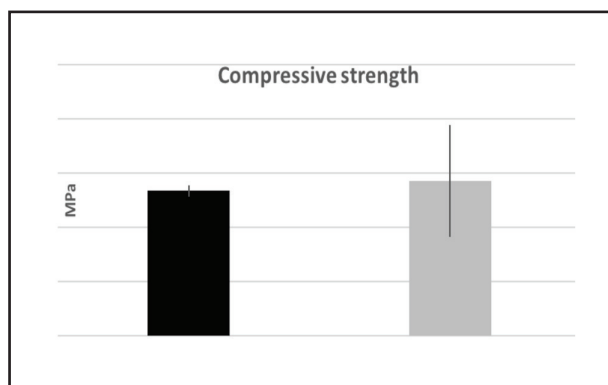


**Figure (1):** TEM images of the sintered silica: (a) before silanization, (b) after silanization together with TiO<sub>2</sub> nanoparticles, at 200 nm magnification.

Results of compressive strength testing, as shown in table (3) and (Figure 2), revealed no statistically significant difference between both composite resin types; the experimental and the control (Nexcomp), where ( $p=0.696$ ). The control (Nexcomp) resin composite showed higher compressive strength mean value ( $143.00 \pm 51.67$ ), when compared to the experimental composite ( $133.60 \pm 5.41$ ).

**Table (3):** Mean and standard deviation (SD) values of compressive strength (CS) (MPa) of both groups.

Variables	Compressive strength	
	Mean	SD
Experimental	133.60 <sup>a</sup>	5.41
Control (Nexcomp)	143.00 <sup>a</sup>	51.67
<i>p-value</i>	<b>0.696 ns</b>	



**Figure (2):** Bar chart representing mean values of compressive strength (CS) of the experimental and control core composites.

### DISCUSSION

The field of resin composites is still open for more achievements<sup>(3,13)</sup>. Nanomaterials are believed to introduce better biological, esthetical and mechanical aspects that could add a lot to resin composites to function in all areas of the mouth even in high stresses bearing areas. This study concerned to prepare experimental composite utilizing the target properties of nanosilica and nanotitanium particles to be used as core material with high compressive strength capable to sustain the chewing and masticatory forces.

For the resin matrix preparation, Bis-GMA was used as a major monomer in the resin structure. It is one of the most traditionally used monomers that produce adequate mechanical properties and low polymerization shrinkage. However, it has high viscosity owing to its high molecular weight and more Hydrogen bonds. Therefore, low molecular weight TEGDMA was added as diluent to lower the viscosity. TEGDMA has been frequently used in dental composites as a functional monomer. It produces high number of double bonds per unit of weight on a flexible backbone and so, improves the conversion process<sup>(1, 14)</sup>.

In the current study, the photo-initiator system was based on camphorquinone, and a tertiary amine; ethyl diethyl amine benzoate. The amine serves as a proton donor that accelerates free radical production. While, camphorquinone has a wide spectrum of absorption wave lengths from “360 – 510 nm” (blue visible light) which is proper with the light emitting diodes (LED)<sup>(14)</sup>.

Typically, dental resin composites consist of four main components; a polymeric matrix, inorganic fillers, a coupling agent and photo initiator system. Silanization of the fillers is always preferred as it provides a crucial link between the matrix phase and the filler phase. In this study 3wt% of trimethoxy silane was used to produce a homogenous distribution of the fillers in the resin matrix by preventing undesirable precipitation and agglomeration. Therefore, adequate wetting was achieved that aimed to improve the overall performance of the material<sup>(9,14)</sup>.

Addition of few drops of acetic acid gradually to obtain a pH of 3-4 helped to increase the hydrolysis rate of the silane coupling agent to improve the stability of the silanol groups. During preparation, the glass beaker was covered with aluminum foil to avoid evaporation of ethanol as well as trimethoxy silane during the stirring process<sup>(9)</sup>.

Sintering was done at 1300 °C that enables forming of clusters that enabled high filler loading without counteracted viscosity of the nanoparticles. In this study the 1 wt% of TiO<sub>2</sub> nanoparticles loading was used according to previous studies which revealed that it positively improves the mechanical properties of the resin composite including microhardness, flexural strength and flexural modulus<sup>(15,16)</sup>.

In the present study, Nexcomp nanohybrid composite was used as control; one of the commercially available dental composites in the Egyptian dental market, of affordable cost and most commonly used as direct and indirect restorative material.

Compressive strength of core materials is an important property as they usually replace the large bulk of tooth structure that resist multidirectional masticatory and para-functional lateral forces. In the present study, there was no significant difference in results of compressive strength test performed for the experimental and control resin composites.

The experimental composite core type revealed lower insignificant difference regarding the mean compressive values (table 3). However, the two tested materials had mean CS values (>100 MPa) higher than the minimum value (50 MPa) accepted for dental amalgam<sup>(12,17)</sup>.

Chemical composition of resin matrix as well as filler particles shape, size, distribution and surface treatment are among the main factors influencing the mechanical properties including the compressive strength of a restoration. The resin matrix in both the experimental and the control share the presence of Bis-GMA and TEGDMA monomers. However, the control resin composite has other monomer types; UDMA and Bis-EMA in the resin matrix, in addition to nanohybrid filler loading (0.04-7µm). On the other hand, the presence of spherical nanosilica (5-15 nm particle range) and TiO<sub>2</sub> nanoparticles (32 nm) in the experimental resin composite with addition of silica clusters allowed for proper filler size distribution as well as high filler loading that reached (85 wt%). Moreover, the addition of spherical shape nanoparticles in the experimental composite have the advantage of increasing the strength of a restoration as stresses are more prone to be concentrated at sharp angles. This may explain the comparable insignificant difference in the compressive strength results of both experimental and control resin composites.

These results are in agreement with other studies that found no statistically significant difference in the compressive strength results of nanofilled and nanohybrid commercial composite types<sup>(18,19)</sup>. Another study found that nanocomposite showed higher compressive strength values than dental amalgam and resin modified glass ionomer<sup>(20)</sup>. From the above results, it could be emphasized the success of the experimental nanocomposite to be used as dental core material in terms of compressive strength performance.



## CONCLUSIONS

Within the limitations of the current study, the following conclusions were drawn:

1. Addition of up to 85 wt% of sintered nanosilica, titanium dioxide and silicon dioxide nanofillers into traditional resin matrix produced a resin composite with comparable compressive strength to that of the commercial composite products commonly used as core material.
2. The experimentally prepared resin composite core material represents a hope for a trustful restoration in the dental field.

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