



## Effects of silane-modified fillers on properties of dental composite resin



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### ABSTRACT

The effect of silanization on the mechanical, chemical, and physical properties of dental composites was investigated. Silica fillers were obtained from colloidal silica solution, Ludox® HS-40 and they were silanized by using 3-methacryloxypropyl trimethoxysilane (MPTMS) in an acidic media. Mineralogical and chemical structures of unsilanized and silanized fillers were determined by using XRD and FT-IR analyses. The modification of unsilanized/silanized fillers were investigated by performing XPS and TGA analyses. The morphological evaluations, surface area, and particle size measurements were performed by using SEM, BET, and Zeta-Sizer, respectively. Eventually, pure and amorphous silica fillers were obtained. Furthermore, the weight percentage of the silane in silica/silane structure was compatible with theoretical values. SEM images, surface area, and particle size measurements showed that agglomeration tendencies of silanized fillers were lower compared to silanized fillers because of the MPTMS addition. Experimental composites (5/10/10/5BisGMA/HEMA/UDMA/TEGDMA resin reinforced with 70 wt% silanized/unsilanized SiO<sub>2</sub>) were fabricated into 4 mm diameter × 6 mm thick discs for compressive strength (CS), angular flexural strength (AFS), curing depth (CD), and polymerization shrinkage (PS) on a 25 × 2 × 2mm rectangular Teflon mold for flexural strength (FS) and modulus of elasticity (E) tests. The curing depth (CD) and degree of polymerization percentage (DP) of composites were determined. Consequently, results showed that mechanical properties and DP of composite resins can be greatly influenced by silanization as a result of the organic matrix-inorganic filler interface bonding formed by silane structures. Despite of these findings, silanization of the SiO<sub>2</sub> was not effected DC and PS values.

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### 1. Introduction

Tooth decays as well as dental diseases negatively affect both the general health status and the appearance of an individual. Even though cavities do not cause life threatening pain, pulpitis pain is important and it disturbs the patient [1]. Therefore, early diagnosis and the treatment of the tooth decays are important [2]. Dental restorative materials are composed of mainly organic resins [3], inorganic fillers [4], and initiator systems [5,6]. Although reinforcing phase systems (fillers) have been used to improve the appearance and mechanical properties of polymeric structures, it is still not possible to use these composite structures in large-scale restoration implementations [7]. Recently, various composite filling materials such as Filtek™ LS (3 M ESPE), Aelite™ LS (Bisco), Kalore™ (GC America), N'Durance™ (Septodont), and Grandio® (VOCO America) are alleged to exhibit low volume shrinkage [8,9]. Some of these materials contain BisGMA as an organic matrix monomer. Along with high rates of fillers in these structures, it is possible for them to display 'low polymerization shrinkage' without adding low molecular weight diluents [10]. Another approach is to decrease the

polymerization shrinkage of methacrylates by using high molecular weight monomers such as dimeric acid derivative [11]. Epoxy based ring opening monomers are also accepted as alternative monomer structures since they decrease shrinkage stresses [12]. Silorane based composites which are members of these monomer systems, can decrease the polymerization shrinkage and polymerization reactions are not inhibited in the presence of oxygen [10]. However, mechanical features of silorane based dental composites are weaker compared to Bis-GMA based resin matrices [13,14]. In this context, the production of composites which are more similar to organic-inorganic phase integrity of the natural teeth, can be achieved by using "silane-based binding agents". These agents also ensure the chemical organic-inorganic matrix interconnection [15,16]. Surface modification of nanoparticles by using silane-based binding agents is a useful way to improve the homogenous dispersion of nanoparticles in various liquids [17,18]. Early studies related to the use of silane-based binding agents were performed by Plueddemann et al. [19]. Then, researchers focused on studies in which they tried to develop the compatibility between particles and the polymer surface and features of composite material by modifying the particles with silane-based binding agents [17].  $\gamma$ -methacryloxypropyltrimethoxysilane (A174) is the most widely used bonding agent since it can efficiently bind to organic and inorganic materials [15]. Recently, the positive effect

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of silanization on mechanical features has been known. Mechanical features of composite structures which are produced by the modification of inorganic particles with A174, are higher compared to composites containing unmodified particles [15,16].

In the present study, 3-methacryloxypropyltriethoxysilane (3-MPTMS) was used as a silane coupling agent in order to functionalize the silica filler. The main objective was to examine the effect of silanization of fillers on the mechanical, chemical, and physical properties of experimental dental composite resins. The surface area of the silica filler is increased by means of silanization and silanized filler could be well distributed in the organic matrix compared to the unsilanized filler. Moreover silane coupling agent also promotes a chemical interaction between the filler and the organic matrix. Such interactions were characterized by using thermo gravimetric analysis (TGA) and X-ray photoelectron spectroscopy (XPS). The results of the mechanical characterization of silanized composites were compared to the mechanical properties of the unsilanized silica reinforced composite. It is expected that mechanical and chemical features of silanized composites are more superior to composites containing unsilanized filler structures depending on the chemical binding between the filler and the organic matrix.

## 2. Materials and methods

In this study, Bis-GMA (%1-5), HEMA (%5-10), UDMA (%5-10), and TEGDMA (%1-5) were used as organic matrices without any modification. Colloidal silica solution, Ludox® HS-40 which has 40 nm particle size, was used as the filler. Besides, 3-methacryloxypropyltrimethoxysilane (MPTMS; A174) was used as the silane agent, which is mostly used in dental composites.

### 2.1. Synthesis of silica filler and silica/silane filler systems

Colloidal silica solution was dried in oven at 80 °C and dried dusts were grinded in a ball mill for a day. Upon milling process, they were sieved through the silica 250 mesh (63 µm) and silica fillers were shortly named as SiO<sub>2</sub>.

In case of silanization studies, proper amount of A174 was added to the 380 mL ethanol:water (3:1) solution in the sealed glass vial, the pH value of the solution was adjusted to 3.5 by acetic acid solution, and

then it was stirred for 30 min. Then, silica was added to this solution while it was vigorously stirred, the solution was stirred on a heater with a magnetic stirrer for 30 min and in ultrasonic water bath for 10 min. After stirring, the solution was refluxed at 80 °C for 3 h. The solution was then filtered by using a vacuum filtration method and it was washed with ethanol/water in order to remove the unreacted silane agents. The resulting solution was dried at 60 °C for 24 h in a vacuum oven. A corresponding flow chart depicts the stepwise synthesis of the silica and silane-functionalized silica in Fig. 1. The rate between the silane amount which was used as the binding agent (X) and the amount of the filler was determined by Arkles and it can be seen below [20].

$$X = A \times f / \omega \quad (1)$$

X: the amount of binding agent (g), f: the amount of filler (g), A: the surface area of the filler (m<sup>2</sup>/g), ω: wetting surface of the silane (314 m<sup>2</sup>/g).

The mineralogical structures of fillers were determined by performing XRD analyses. 2θ area was scanned by 0.02° distances and 0.5 s/min. X-rays which were required for the scanning, were obtained by using Cu-Kα radiation (1.54059 Å) and parabolic filter in MiniFlex 300/600, and RIGAKU device by using 40 kV and 15 mA. Chemical structures of fillers were examined by using FT-IR (Spectrum One, PERKIN ELMER) device at room temperature, at 400–4000 cm<sup>-1</sup> wave length and with 4 cm<sup>-1</sup> solubility. Silanized silica fillers were analyzed by X-ray photoelectron spectroscopy (XPS) in order to determine binding properties of the silica and 3-MPTMS components. XPS analyses were performed by using EA 300 and SPECS devices at Al monochromatic mode and 57 W power.

Relative amounts of organic and inorganic structures which were presented in inorganic/organic composite materials can be easily determined by using thermogravimetric analysis methods [21]. In this study, TGA analyses were performed in order to determine the amount of A174 molecules which were used to modify the filler. Thermal analyses were performed by using STA 409 PC/PG, NETZSCH device in the inert nitrogen atmosphere, at between 30 °C and 850 °C, and with the heating speed of 1 °C/min. In this current study, surface morphology of silanized/unsilanized filler systems were examined by using the scanning electron microscope (SEM, JSM 5600, JEOL). Samples were placed in an aluminum stub by spraying on the double-sided tape. Then,

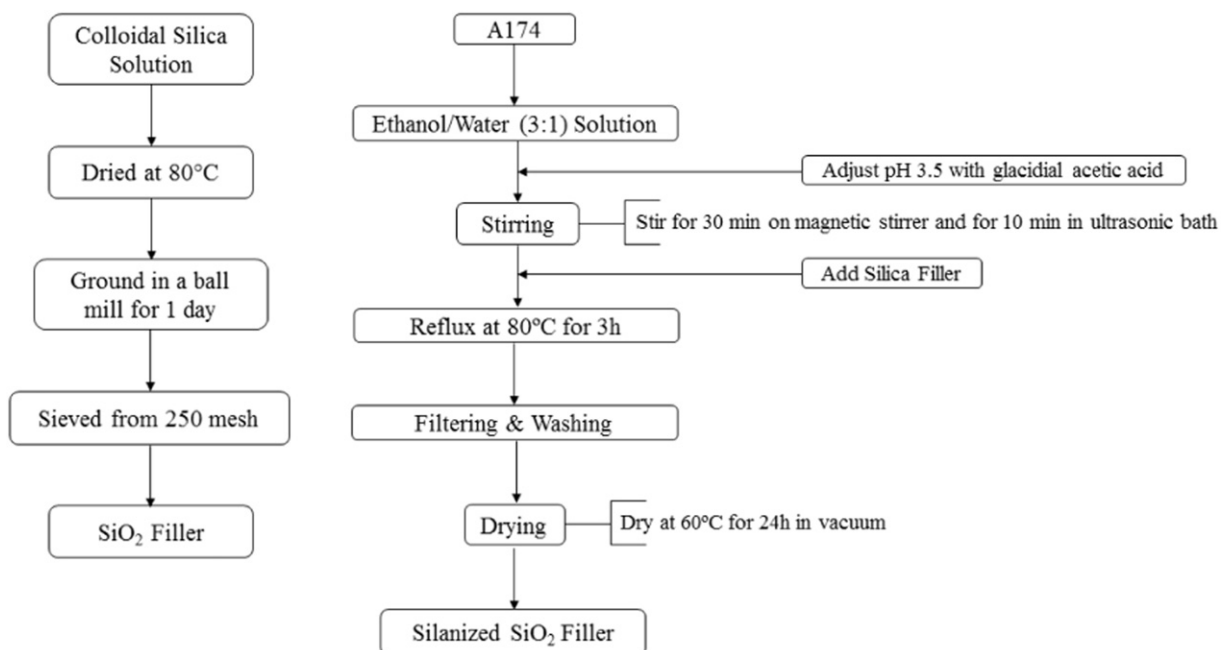


Fig. 1. Process flow chart of silica and silanized silica filler.

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