Performance of Electrospun PMMA-Silica Nanofiber as Reinforced Material in Dental Composite Restoration

Nina Djustiana[1,2], Yanwar Faza[1,2], Nadiya Sudiyasari[3], Annisa Tamyra Firdaus[3], Kosterman Usri[1,2], Arief Cahyanto[1,2]*

1. Department of Dental Materials Science and Technology, Faculty of Dentistry, Universitas Padjadjaran, Indonesia.
2. Oral Biomaterials Study Centre, Faculty of Dentistry, Universitas Padjadjaran, Indonesia.
3. Graduate Dental Student of Faculty of Dentistry, Universitas Padjadjaran, Indonesia.

Abstract

The objective of this study was to investigate the morphology of the electrospun PMMA-silica with varying amounts of silica (0 wt%, 1 wt%, 2 wt%, and 5 wt%) and to evaluate the flexural strength of dental composite reinforced with polymethyl methacrylate (PMMA)-silica nanofibers (1 wt% of silica content) compare with PMMA nanofiber as a control. The PMMA–silica nanofiber was synthesized by the electrospinning process. Later, electrospun PMMA-silica nanofiber was impregnated with methacrylate of BisGMA/TEGDMA (50/50 mass ratio) to prepare dental composite. Scanning Electron Microscopy (SEM) results in the morphology of the PMMA-silica nanofiber were observed with the addition of 1 wt% silica. Along with more additional of silica, the morphology became poor due to irregularities and beads. The XRD patterns revealed that the peak of silica was present, however, the pattern was dominated by the PMMA. The flexural strength of PMMA-silica nanofiber dental composite was 132.74 ± 20.70 MPa and PMMA nanofiber dental composite was 128.99 ± 12.60 MPa. In conclusion, there were no statistically significant in flexural strength values between PMMA-silica nanofiber dental composite and PMMA nanofiber dental composite (p-value > 0.05).

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Introduction

Dental composite is a resin-based restoration that frequently used due to its excellent aesthetic properties.[1-5] It commonly consists of ceramic filler and methacrylate-based resin. Silica is often used as particle filler of dental composite, meanwhile, 2,2-bis-[4-(methacryloxypropoxy)-phenyl]-propane (BIS-GMA) and tri (ethylene glycol) dimethacrylate (TEGDMA) are occasionally chosen as the resin matrix[6-7]. Based upon plenty of studies, long periods of services of the dental composite is still questionable due to wear or loss of anatomical form after 18 months.[6,8] Along so many investigations, it reveals that filler particles are contributed to the deterioration of dental composite caused by its angulated and irregular shape lead to be a stress raiser.[6,7,9,10]

Electrospun nanofiber possesses desire morphological and mechanical properties including large aspect ratio, small fiber diameter, and high strength. It can be a reinforcing material because of its “bridging” mechanism against the microcrack that passing through the matrix by remaining intact across the crack planes.[1,7,10] The polymethyl methacrylate (PMMA) is a polymer that is commonly used in dentistry, PMMA has been chosen based on its aesthetics, fabrication method, and economic point of view.[11,12] Previous research already used PMMA, combination with PAN in nanoscale fiber, as a filler to substitute filler particles of dental composite. It exposed that the electrospun nanofiber can elevate the flexural strength by 47%.[13] However, there is no experiment to use the PMMA with silica in the nanofiber form as the filler of dental composite. The propose of this study is to synthesis PMMA-silica nanofiber with varying amounts of silica (0 wt%, 1 wt %, 2 wt%,

Corresponding author:
Arief Cahyanto,
Department of Dental Materials Science and Technology, Faculty of Dentistry, Universitas Padjadjaran, Jalan Sekeloa Selatan 1 Bandung, 40134, West Java, Indonesia.
E-mail: arief.cahyanto@fkg.unpad.ac.id

and 5 wt%) then fabricate the dental composite using nanofiber of PMMA-silica (1 wt%) then evaluate its flexural strength.

Materials and methods

Polymethyl methacrylate (PMMA), silica nanoparticles, bisphenol A glycidylmethacrylate (BisGMA), triethylene glycol dimethacrylate (TEGDMA), 2-diethylmethacrylate (DMAEMA) camphorquinone (CQ) were purchased from Sigma Aldrich, Singapore. Acetone was purchased from a chemical local store Bratachem, Bandung, Indonesia.

This study started by produced electrospun PMMA-silica fiber with electrospinning process. PMMA particles (0.1 g) were dissolved into acetone (4.9 g) then stir for 30 minutes to obtain a homogeneous solution. Subsequently, the amount of silica was added into the solution under continuous stirring for about 24 h. Four groups of specimens with different amounts of silica were fabricated, which contained 0 wt%, 1 wt%, 2 wt%, and 5 wt%. Electrospinning solution was loaded in a 5 ml syringe which will be connected to the positive electrode of high voltage power supply. Metallic collector, wrapped by aluminum foil, was connected to the negative electrode. The experimental setup was conducted with the following situation; 1 ml/h flowing rate, 13 kV electric charge, 12 cm distance from the needle of syringe to the collector and 50% humidity. The morphology and diameter of electrospun PMMA-silica were evaluated using scanning electron microscopy (JEOL JSM-6360LV FESEM, Tokyo, Japan). The specimens for XRD evaluation were crushed into fine powders. The powders were characterized using XRD (D8 Advance Bruker, Karlsruhe, Germany) with a CuKα operated at 40 kV of tube voltage and 40 mA of tube current.

PMMA-silica fiber was carefully cut into pieces with size 2 mm x 25 mm, along fiber direction, in order to proceed by a three-point bending test. Dental composites were produced by laminating the fiber pieces into resin matrix (BisGMA 50 wt%, TEGDMA 50 wt%, CQ 0.5 wt%, and DMAEMA 1 wt%). The vibrator was used to diminish the trapped air bubbles inside the dental composite. Throughout specimens were light-cured for 1 minute then stored at room temperature for 48 h. In the final step, specimens were polished before test. Three-point bending test was done using a universal testing machine (Llyod Instruments Ltd., West Sussex, UK) with a crosshead speed of 0.5 mm/min and a span of 20 mm. Acquired data were statistically analyzed using independent t-test with a significant level that was considered at p-values ≤ 0.05.

Results

Figure 1 shows SEM images of PMMA-silica nanofiber with various amounts of silica content (0 wt%, 1 wt%, 2 wt%, and 5 wt%). The diameter of fiber with no silica content (0 wt%), in figure 1 (a), ranges from 30 – 496 nm with the highest frequency is around 300 nm. In this figure, fiber looks smooth with no beads shown on the image. Figure 1 (b), silica is shown with a diameter of 2 nm while the PMMA nanofiber exhibit more varies diameter size of fiber than the previous specimen. The fibers are ranging from 124 – 1214 nm.

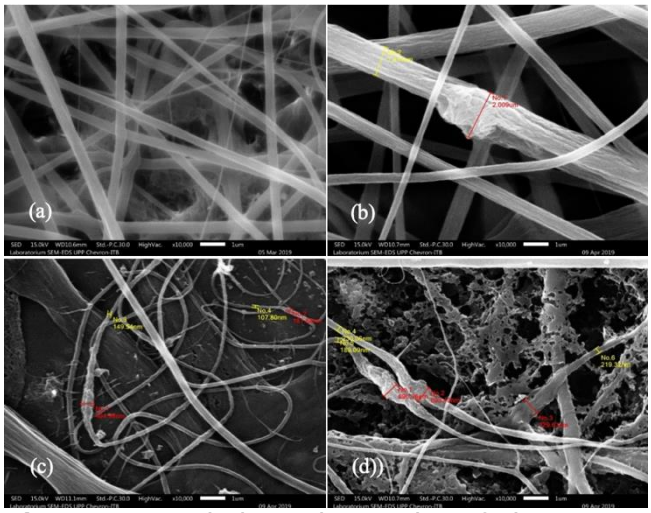

Figure 1. Morphology observation of electrospun PMMA-silica nanofiber by SEM images (a) PMMA nanofiber with 0 wt% of silica ; (b) PMMA-silica nanofiber with 1 wt% of silica ; (c) PMMA-silica nanofiber with 2 wt% of silica and (d) PMMA-silica nanofiber with 5 wt% of silica.

Figure 1 (c) has the most various diameter size of fiber. It ranges from 19 -1162 nm. Silica particles are covered by PMMA fiber with a diameter size, 494 nm. An additional 2 wt% silica produces instability of solution spinning thus create more branching of fiber. Subsequently, figure 1 (d) shown PMMA unevaporated

completely thus display irregular morphology surround the fibers. Diameter size is also different which is ranging from 60 - 729 nm.

X-Ray Diffraction results (Fig. 2) at glance, It shows similar patterns between specimens without silica (a) and with silica content (b, c, and d). However, subsequent analysis using Match! software, the different peaks are observed at 26.65° for PMMA-silica nanofiber.[14] The peaks indicate the presence of quartz silica.

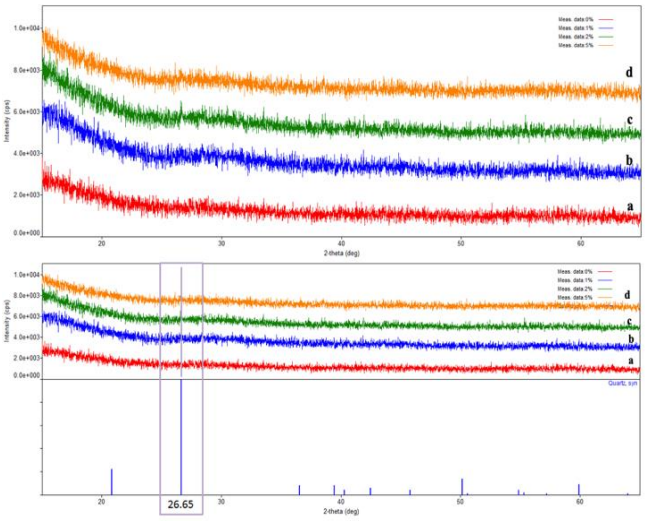

Figure 2. XRD pattern results of PMMA-silica nanofber with different silica content (a) 0%, (b) 1 %, (c) 2% and (d) 5%.

Table 1 summarizes the Fiber PMMA as reinforces material contributes to obtaining 128.99 MPa of flexural strength of the dental composite. On the other hand, fiber PMMA-silica fiber result of 132.74 MPa.

Specimens	Flexural strength value (MPa)	SD	*p*-value
Fiber PMMA dental composite	128.99	12.60	> 0.05
Fiber PMMA-silica dental composite	132.74	20.70	

Table 1. Flexural strength of fiber PMMA dental composite and fiber PMMA-silica dental composite (n=5).

Flexural strength of fiber PMMA dental composite compare to fiber PMMA-silica dental composite is not significantly different (p-value > 0.05).

Discussion

Morphology of PMMA- silica nanofiber with varying amounts of silica show different features and shapes of acquired fiber (Fig. 1). The similar diameter size of fibers is shown in the specimen with no silica content. It due to the solution is more homogenous than others (Fig. 1 (a)). The diameter of fiber also tends to have a lower size (30 - 496 nm) due to the lower concentration of the solution.[15] Meanwhile, in Fig. 1 (b), the particle of silica is present on the surface of nanofiber and exhibit various diameter size of fiber. The addition of silica may increase the viscosity of the solution.[7,15] Therefore, it may increase the diameter size of the fiber up to 1214 nm. In PMMA-silica fiber with 2 wt% of silica content (Fig. 1 (c)), irregularities and beads have existed on the surface of the nanofiber. The diameters of nanofiber start becoming non-uniform and show the poor morphology due to the higher viscosity. At last, PMMA-silica nanofiber with 5 wt% silica content reveals more irregularities and non-uniform of nanofibers. These problems also indicate the failure of electrospinning to overcome high solution viscosity.[15]

On the electrospinning process, it is crucial to achieving a balance between viscoelastic force, surface tension and electrostatic repulsion.[15,16] Since 13 kV is applied, the electric field is enough to provide electrostatic repulsion and maintain the surface tension and viscoelastic force in lower concentration solutions. It favors the stabilization of the liquid jet and produces the smooth surface morphology of nanofiber. However, the voltage is not enough for higher viscosity thus generate instability and the resultant nanofiber has poor surface morphology (irregular and non-uniform). Meantime, it assumes the uneven fibers appear may also due to poor dispersity of the silica in the solution.

Dental composite reinforced with PMMA nanofiber and PMMA-silica nanofiber results 128.99 MPa and 132.74 MPa, consecutively. Based on statistical analysis, there are no statistically significant between them (p-value > 0.05). The study result of PMMA-based nanofiber as reinforcing material of dental composite is similar to the result performed by Sun et al.[13] Their study using PAN-PMMA nanofiber increase the flexural strength of dental composite (130 MPa) compare to neat resin (88 MPa); BisGMA/TEGDMA (50:50). It is believed that polymer PMMA can form an *In situ* nano-interface (IPN) structure that provides good

interfacial adhesion between nanofiber and the resin matrix. At the same time, nanofibers can improve the mechanical properties of BisGMA resin due to their ultrahigh interfacial area.[9,13]

Conclusions

Electrospun of PMMA-silica were prepared by the electrospinning process shown better morphology with an additional 1 wt% of silica. The morphology becomes poor along with more additional silica (2 wt% and 5 wt%). There is no statistically significant between the flexural strength of PMMA-silica nanofiber dental composite compare to PMMA nanofiber dental composite (p-value > 0.05).

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Declaration of Interest

The authors declared that there are no conflicts of interest.

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