Original Article

Flexural Strength of Various Sizes and Weight of Silane-treated Alumina Reinforced Heat-polymerized Acrylic Resins

Duangjai Uraivichaikul[1], **Issarawan Boonsiri**[1], **Kamolporn Wattanasirmkit**[2]

[1]Department of Prosthodontics, College of Dental Medicine, Rangsit University, Pathum Thani, Thailand

[2]Department of Prosthodontics, Faculty of Dentistry, Chulalongkorn University, Bangkok, Thailand

Abstract

The objective of this study was to evaluate the effects of 2.5%, 5%, and 10% by weight silane-treated nano-alumina and alumina reinforcement on the flexural strength of heat-polymerized acrylic resins. Seventy rectangular (65x10x3 mm^3) (ISO standard 1567) heat-polymerized acrylic resins were fabricated and divided into seven groups according to the size and weight % of aluminum oxide reinforcement (n=10). The control group was an intact heat-polymerized acrylic resin group. The six experimental groups were reinforced with 2.5%, 5%, and 10% by weight silane-treated nano-aluminum oxide (20 nm) and aluminum oxide particles (50-70 μm). The specimens were tested for flexural strength using a three-point bending test. Independent *t*-test, One-way ANOVA and Tukey Honestly Significant Difference (HSD) were used for statistical analysis. The flexural strength of 10% by weight of silane-treated nano-alumina and silane-treated alumina reinforced groups (Groups NA3, A3) (111.36, 110.45 MPa) significantly increased compared to the control group (Group C) (99.25 MPa) ($p<0.05$), even though the flexural strength of 2.5% and 5% by weight of silane-treated nano-alumina and silane-treated alumina reinforced groups (Groups NA1, NA2, A1, A2) (101.00, 105.34, 99.27, 104.42 MPa) did not increase significantly compared to the control ($p>0.05$). Group NA3 showed the highest flexural strength compared to the control ($p<0.05$). Changing the percentage by weight of silane-treated aluminum oxide reinforcement affected the flexural strength of heat-polymerized acrylic resins. The flexural strength increased when 10% by weight of silane-treated nano-alumina and alumina fillers were added. Both silaned-treated micro-sized and silaned-treated nano-sized alumina can be used to reinforce the flexural strength of heat-polymerized acrylic resins due to resembling the effect of increase in flexural strength.

Keywords: Acrylic resins, Flexural strength, Nano-alumina, Reinforcement, Silane-treated alumina

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Correspondence to:
Duangja Uraivichaikul, Department of Prosthodontics, College of Dental Medicine, Rangsit University, Pathum Thani, 12000, Thailand.
Tel: 02-521-5031, 080-068-4400 Email: Duangjai.j@rsu.ac.th

Introduction

Polymethyl methacrylate (PMMA) is the most commonly used material to fabricate denture bases.[1] The popularity of PMMA is mainly due to various superior properties such as biocompatibility, ease of processing, stability, low cost and esthetic properties.[2] However, several inferior physical and mechanical properties have prevented PMMA from becoming an ideal denture base material. It has low thermal conductivity, a high coefficient of thermal expansion, relatively low modulus of elasticity, and its brittleness makes the material more susceptible to clinical failure.[3] A variety of physical properties can be used to assess the strength of denture materials. The most common tests are impact strength, the ability of a material to resist a sudden high level force or "shock," flexural strength, and the force needed to deform the material to fracture or irreversible yield. One of the desirable properties in denture base material is high impact stress, due to the risk of fracture that could happen if the patient drops their dentures. In order to achieve a longer clinical denture life, increasing the flexural strength may help resist torsional forces which can be found in the function of an acrylic resin denture base.[4]

Denture base fracture is considered the primary mode of clinical failure; up to 68 % of acrylic resin dentures break within a few years of fabrication.[5] Denture wearers have to make a new denture or spend more money on repairing the old one but it does not last long. Therefore, to extend the longevity of acrylic resin dentures, several methods have been introduced to reinforce the material used for denture base fabrication, such as chemical modification to prepare high impact resin and mechanical reinforcement with glass fibers, sapphire whiskers, aramid fibers, carbon fibers, nylon polyethylene fibers and zirconia.[6] An addition of glass fiber showed a higher impact strength and improved the flexural strength of PMMA.[7] The current meta-analysis showed that the reinforcement of PMMA can considerably enhance both flexural strength and impact strength.[8] Titanium, zirconia and nano-zirconia reinforcement also contributed to an increase in flexural and impact strength of PMMA.[9-11] A recent study showed that the reinforcement of nano-zirconia may improve the transverse strength of a repaired acrylic denture base.[12] The incorporation of 0.75%-ZrO_2 or 0.25% -SiO_2 into repair resin can increase the repair strength.[13] However, the use of metal powder filler has caused the dentures to be unaesthetic. Ideally, metal powder should be incorporated into the resin and increase the strength without affecting other properties.

Furthermore, natural fibers can also be reinforced to increase the mechanical properties of PMMA. These natural fibers are plant-based and lignocellulose in nature. They are composed of lignin, pectin, cellulose, hemicelluloses and waxy substances. Natural fibers are placed in the weak areas of the denture and 90° to the fracture line. The fibers become stronger when they are placed in a unidirectional direction.[14] A study conducted by Oleiwi *et al.*, demonstrated that pure PMMA specimens had a lower compressive strength than that of PMMA specimens reinforced with miswak or bamboo fibers.[15]

One of the materials that is commonly found in dental settings is aluminum oxide, which is commonly referred to as alumina. It possesses strong ionic interatomic bonding, giving rise to various desirable material characteristics. It can exist in several crystalline phases, which all revert to the most stable hexagonal alpha phase at elevated temperatures. This is the phase of particular interest for structural applications. Alpha phase alumina is the strongest and stiffest of the oxide ceramics. It has more hardness, excellent dielectric properties, refractoriness, and good thermal properties make it the material of choice for a wide range of applications in dentistry.[6,16] Antimicrobial properties are considered a desirable characteristic that nanoparticles should possess. Aluminum oxide nanoparticles have a wide range of applications in the industry, and are also known to possess antimicrobial properties in a dental aspect. Alumina showed a mild bacterial growth-inhibitory effect, but only at very high concentrations.[17] Other research showed that the antimicrobial properties of nanoparticles containing formulations were increased.[18] The addition of 5% by weight aluminum oxide nanoparticles to acrylic resin also improved the thermal properties and transverse strength of acrylic resin, and at the same time decreased its water sorption and solubility.[10] On the other hand, there was an increase in surface roughness of the acrylic resin, yet the surface roughness did not significantly change even though the concentration of aluminum oxide nanoparticles was increased.[19]

In addition, the reinforced particle shape affects the mechanical properties of PMMA. Elongated particles are more efficient in improving the properties of acrylic resin than spherical particles.[20] There were various studies presenting the effect of particle size and percentage of concentration of particles on the flexural strength of acrylic resin. An addition of 10% by weight and 15% by weight of alumina powder to heat cure acrylic resin improved the flexural strength.[21] Ten percent by weight of treated alumina particles sized 18-23 µm. resulted in a 23.86 % increase in flexural strength.[22] One percent by weight of treated nanofillers of alumina increased 24 % of flexural strength.[23] Five percent by weight of 5-22 µm. untreated alumina particles improved flexural strength by 13.99 %.[24] One percent and 1.5% by weight nano-Al_2O_3 increased the flexural strength of the repaired denture.[25] Nanofillers are more effective than microfillers at lower concentrations. Moreover, studies demonstrated that the use of alumina particles treated with silane coupling agent leads to a significant increase in properties of acrylic resin as compared to untreated particles, as it improves the surface bonding of filler and resin matrix. Previously, a study presented that 10% by weight of treated particles with silane coupling agent 18-23 µm particles led to a 23.86 % increase in flexural strength whereas non-silanized resulted in a 5.46 % increase in the strength.[22]

Therefore, this study aimed to evaluate the effect of 2.5%, 5%, and 10% by weight of silane-treated nano-aluminum oxide (20 nm.) and silane-treated aluminum oxide (50-70 µm) reinforcement on the flexural strength of heat-polymerized acrylic resins. The null hypothesis was that there is no significant difference in flexural strength of heat-polymerized acrylic between micro-sized and nano-sized aluminum oxide reinforcement. Also, there is no significant difference in flexural strength of heat-polymerized acrylic among different weight percentages of aluminum oxide particles. Lastly, there was no significant difference in flexural strength of heat-polymerized acrylic amongst the various sizes, volumes of aluminum filler and the control group.

Materials and methods

Seventy ($65*10*3\ mm^3$)[26] (ISO standard 1567) PMMA specimens were fabricated from heat polymerization (Meliodent Heat Cure, KULZER MITSUI Chemicals group, Heraeus Kulzer Gmbh, Tokyo, Japan). All specimens were prepared using a heavy-body condensation silicone mold made of rectangular custom-made acrylic blocks. Pink wax was melted and placed into the silicone mold, where glass slides were used to control the thickness of the rectangular wax pieces. The rectangular wax pieces were invested in a metal flask with dental stone. After setting the dental stone, the flasks were placed in a scalding unit for wax boil-out (100°C, 5 minutes), leaving the rectangular shaped mold cavity in the dental stone, which was used as a matrix for the fabrication of specimens. A separating medium was applied to the stone mold.

Table 1 *Materials used in this study*

Materials	Use of materials	Compositions	Manufacturer
Heat cured acrylic resins	Denture base processing	Powder: 95% PMMA, 4% plasticizer, 1% benzoyl peroxide Liquid: 90% MMA, 10% dimethacrylate, catalyst	Meliodent KULZER MITSUI Chemicals group, Heraeus Kulzer Gmbh, Tokyo, Japan
320-grit silicon carbide paper	Finishing and polishing	C 0.07%, SiC 93.9%, Fe_2O_3 0.64%, Si 0.79%, Al_2O_3 0.25%, CaO 0.20%, SiO_2 4.11%	TOA Paint Public Company Limited, Thailand
Silane coupling agent (MPS)	Silanization	1% 3- methacryloxypropyl trimethoxysilane ethanol/water-based solvent, acetic acid	KBM 503, Shin-Etsu Chemical, Tokyo, Japan
Alumina particles with diameters ranged from 50-70 µm	Reinforcement	Al_2O_3 95%, SiO_2 0.72% Fe_2O_3 1.00%, TiO_2 3.00% CaO 0.30%	Kepler International, Thailand
Nano-alumina particles with diameter 20 nm	Reinforcement	Al_2O_3, gamma, 99%, 20 nm	US Research Nanomaterials, Inc., Houston USA

1. Specimen preparation

All heat-polymerized PMMA specimens were divided into seven groups: one control group (intact heat polymerized specimens) and six experimental groups (addition of nano-alumina and alumina powder) as shown in Table 2.

Table 2 Experimental groups and coding according to reinforcement material used for reinforcing process

Group code	Reinforcement material used for reinforcing process
C	Intact heat-polymerized acrylic resin specimens (control)
NA1	Heat-polymerized acrylic resin reinforced with 2.5% by weight silane-treated nano-aluminum oxide particles (20 nm)
NA2	Heat-polymerized acrylic resin reinforced with 5% by weight silane- treated nano-aluminum oxide particles (20 nm)
NA3	Heat-polymerized acrylic resin reinforced with 10% by weight silane- treated nano-aluminum oxide particles (20 nm)
A1	Heat-polymerized acrylic resin reinforced with 2.5% by weight silane- treated aluminum oxide particles (50-70 μm)
A2	Heat-polymerized acrylic resin reinforced with 5% by weight silane- treated aluminum oxide particles (50-70 μm)
A3	Heat-polymerized acrylic resin reinforced with 10% by weight silane- treated aluminum oxide particles (50-70 μm)

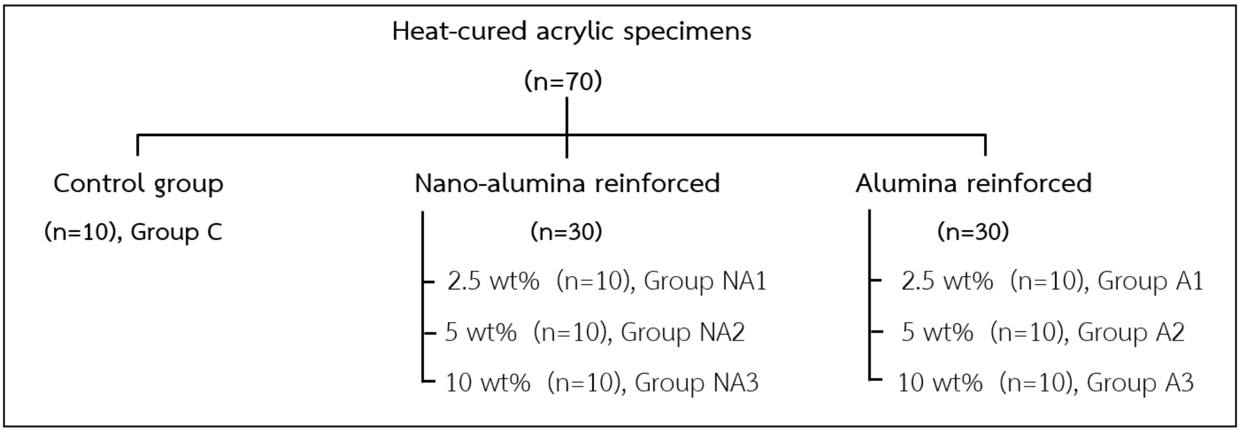

***Figure 1** Sample groups*

Aluminum oxide particles (50-70 μm) (Kepler international co., LTD Thailand) and nano-aluminum oxide particles (20 nm) (US Research Nanomaterials, Inc., Houston, USA) were pre-weighed into 2.5%, 5%, and 10% by weight of heat-polymerized acrylic resin particles using an electronic weighing machine. Then, 0.1% silane coupling agent MPS (3-methacryloxy propyl trimethoxysilane) (KBM 503, Shin-Etsu Chemical, Tokyo, Japan) was measured using a micropipette (10-100 μl size, SCILAB) according to Arkle's equation [27]:

$$\text{Amount of silane (g)} = \frac{\text{Amount of filler (g)} \times \text{surface area (m}^2\text{/g)}}{\text{Minimum coating area of silane coupling agent (m}^2\text{/g)}}$$

Silane was used to coat the preweighed aluminum oxide particles using a microbrush and a one-minute waiting drying time. Then, preweighed 2.5%, 5%, and 10% by weight aluminum oxide particles and nano-aluminum oxide powder with resin polymer powder (50 mg) was mixed using a magnetic stirrer (IKA C-MAG HS7), in order to achieve an equal distribution of particles and uniform consistency.[28] Packing and processing of specimens were done according to the manufacturer's instructions. The upper and lower flasks were closed and maintained under

200 Ib of compression for 30 minutes. The flasks were removed from the hydraulic pressure and cooled over the bench for 150 minutes. The curing procedure was processed by placing the flasks in the water bath at 71°C for nine hours.[3] Flasks were allowed to cool to room temperature overnight before opening and deflasking. The specimens were removed from the mold, finished, and polished with 320-grit silicon carbide paper (TOA Paint Public Company Limited, Thailand) using a polishing machine (Whip Mix Corporation, Kentucky, USA). The polished specimens of each group are shown in Figure 2. The specimens were later stored in an incubator at 37°C for two days before flexural strength testing.[29]

Figure 2 *Polished heat-polymerized acrylic resin specimen of each group (65*10*3 mm^3)*

2. Flexural strength testing

Before flexural strength testing, the thickness, width, and length of each specimen were examined for accuracy by using a digital micrometer (minimum reading: 0.001 mm, Digimatic Micrometer Mitutoyo Corp., Kanagawa, Japan). Hence, the midpoint in length of each specimen was determined and marked. The flexural strength of the specimens were determined by using a three-point bending testing device in a universal testing machine (EZ test, Shimadzu, Japan). The device is composed of a loading wedge and a pair of adjustable supporting wedges placed 50 mm. apart. The specimens were centered on the device in such a way that the loading wedge, (with a set crosshead speed of 5 mm/min), engaged the center of the upper surface of the specimens. Specimens were loaded until fracture occurred. Flexural strength was calculated using the following equation [29]:

$$S = 3Pl/2bd^2$$

S is the flexural strength (N/mm^2), P is the fracture load (N), l is the distance between the supporting wedges (mm), b is the specimen width (mm), and d is the specimen thickness (mm).

3. Visual surface analysis

A scanning electron microscope (FEI QUANTA 200, FEI Company, Oregon, USA) was used to visualize the surface topography and examine the filler distribution in the polymer matrix of the control and experimental specimens. It was also used to analyze the mode of failure of each experimental specimen. Definitions of each mode of failure is described in the following sentences: Adhesive failure means that fracture occurs between the interface of silane-treated aluminum oxide particles and PMMA. Cohesive failure means that fracture occurs between PMMA and PMMA. Mixture of adhesive and cohesive failure means that fracture occurs between the interface of the silane-treated aluminum oxide particles and PMMA (adhesive failure) and between PMMA and PMMA (cohesive failure). One sample from each group was coated with gold by Gold Coater Jeol Model JFC-1200 and visualized at 200-1500x magnification. An attempt was made to focus on a presenting area and adjustment of the higher magnification while remaining on the same area was also done.

4. Statistical analysis

Data analysis was performed using SPSS software version 23, SPSS Inc, IRM corp, Chicago IL, USA at 95% confidence of level. A *p*-value ≤ 0.05 was significantly considered. After obtaining results from the experiment, it was used to test for normality using the Sharpiro-Wilk test as the degree of freedom of each group which was less than 50. The results were well-modeled by normal distribution and parametric statistical tests were done.

The first hypothesis focused on a single independent variable: size of particles, which was further divided into two sets of data, including the nano-aluminum oxide (20 nm) reinforced group and the aluminum oxide (50-70 μm) reinforced group. The single dependent variable was the value of flexural strength. An independent *t*-test was done in order to compare means of flexural strength between the two groups. An independent *t*-test is an inferential statistical test that determines whether there was a statistically significant difference between the means in two unrelated groups.

The second and third hypotheses were analyzed using One-way ANOVA, as there were seven sets of data, a single independent variable, and one dependent variable. Afterwards, the Post-Hoc or Multiple Comparison test was used to determine which of the paired means amongst a set of means was significantly different from the rest. Tukey's Honestly Significant Difference (HSD) test was chosen for pair-wise comparison due to the equal number of sample sizes in each set of data.

Results

As shown in Figure 3, in the investigation of the effect of the size of aluminum oxide particle reinforcement on the flexural strength of heat-polymerized acrylic resin, the null hypothesis was accepted. Changing the size of reinforced alumina, between nano-aluminum oxide fillers (20 nm) and aluminum oxide fillers (50-70 μm), did not significantly affect the flexural strength of heat-polymerized acrylic resin. The mean flexural strength of nano-alumina and alumina filler reinforcement were 106.20 MPa and 104.71 MPa, respectively. Although the mean flexural strength of the nano-aluminum oxide reinforced groups (Groups NA1, NA2, NA3) was higher than that of the aluminum oxide reinforced groups (Groups A1, A2, A3) with a mean difference of 1.49 MPa, the difference was not shown to be statistically significant (*p*=0.484)

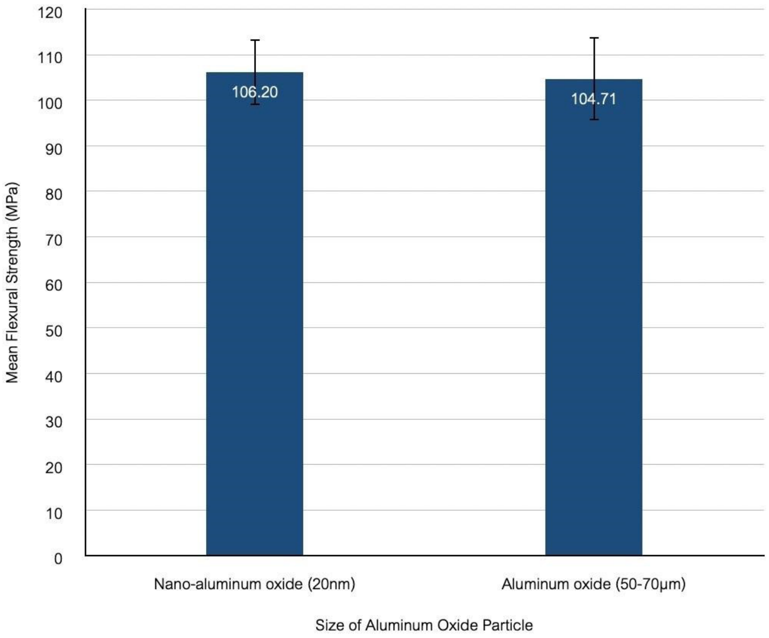

Figure 3 The effect of size of aluminum oxide particle reinforcement on the flexural strength of heat-polymerized acrylic resin

As shown in Table 3, in investigating the effect of weight % of aluminum oxide particle reinforcement on the flexural strength of heat-polymerized acrylic resin, the alternate hypothesis was accepted. There was a significant difference between the flexural strengths of heat-polymerized acrylic resin that resulted from different weight % of fillers reinforced. The mean flexural strength of 10% particle reinforced group (Group NA3, A3) was 5.57 MPa and 10.77 MPa higher than that of the 5% and 2.5% particle reinforced groups (Group NA2, A2 and Group NA1, A1) respectively, which appeared to be statistically significant (*p*=0.038, *p*=0.000). Although the mean flexuralstrength of the 5% particle reinforced group (Group NA2, A2) was 5.20 MPa higher than that of the 2.5% particle reinforced group (Group NA1, A1), the difference was not statistically significant (*p*=0.056).

As shown in Table 4, in the investigation of the effect of size, weight %, and absence of aluminum oxide particle reinforcement on the flexural strength of heat-polymerized acrylic resin, the alternate hypothesis was accepted. There was a significant difference between the flexural strengths of heat-polymerized acrylic resin that resulted from various sizes, weight % of aluminum filler reinforced and control groups. The highest flexural strength of heat-polymerized acrylic resin was presented in 10% by weight of the nano-aluminum oxide group (Group NA3), followed by 10% by weight of the aluminum oxide group (Group A3), with a mean flexural strength of 111.36 MPa and 110.45 MPa. The mean flexural strength of 10% by weight of the nano-aluminum reinforced group (Group A3) was higher than 2.5% by weight of the nano-aluminum oxide (Group NA1), 2.5% by weight of the aluminum oxide (Group A1) and the control group (Group C) by 10.36 MPa, 12.09 MPa and 12.11 MPa, respectively. Likewise, the flexural strength of 10% by weight of the aluminum oxide reinforced group (Group A3) was higher than 2.5% by weight of the nano-aluminum reinforced group (Group NA1), 2.5% by weight of the aluminum oxide (Group A1) and the control group (Group C) by 9.45 MPa, 11.18 MPa and 11.19 MPa, respectively. The mean flexural strengths of both 10% by weight reinforced groups (Groups NA3, A3) were significantly higher than both 2.5% by weight of the reinforced groups (Groups NA1, A1) and the control group (Group C) (*p*=0.018, *p*=0.03). However, the mean flexural strengths of both groups (Groups NA3, A3) were not significantly different from each other, with a mean difference of 0.91 MPa (*p*=1.000). The lowest flexural strength was presented in the control group (Group C), followed by 2.5% by weight of the aluminum oxide group (Group A1), and 2.5% by weight of the nano aluminum oxide group (Group NA1), with a mean flexural strength of 99.25 MPa, 99.27 MPa, and 101.00 MPa, respectively. The aforesaid groups did not show a statistically significant difference among each other (*p*=1.000, *p*=0.997). In addition, the flexural strength of 5% by weight of the nano-aluminum and 5% by weight of the aluminum oxide reinforced groups (Group NA2, A2) were not significantly different from the control group nor any of the five experimental groups.

As shown in Figure 4, results of the SEM study showed that the aluminum oxide particles in all reinforced specimens were widely and evenly distributed within the resin matrix. The presence of reinforced particles also increased as the weight percentage of reinforcement increased. However, the SEM images showed that the shape of aluminum oxide particles varied between the nano-sized particles (NA1, NA2, NA3) and micro-sized particles (A1, A2, A3). The micro-sized aluminum oxide particles were irregular in shape, yet the nano-sized aluminum oxide particles were spherical in shape. SEM images shown in Figure 5 were taken from the fractured surface of nano-aluminum oxide reinforced specimens. The aforesaid images revealed that a mode of failure for the specimens was a mixture of cohesive and adhesive failures. Fracture occurred between the interface of silane-treated aluminum oxide particles and PMMA (adhesive failure) and between PMMA and PMMA (cohesive failure). Mode of failure of each group of all specimens was presented in Table 5. A control group (Group C) experienced 100 % cohesive failure, whereas the other experimental groups (Groups A1, A2, A3, NA1, NA2, and NA3) experienced a mixture of 100 % cohesive and 100 % adhesive failures.

Table 3 *The effect of percentage by weight of aluminum oxide particle reinforcement on the flexural strength of heat-polymerized acrylic resin*

Weight % of aluminum oxide particle [I]	Mean flexural strength (MPa)	Standard deviation	Mean difference (*p*-value) [J]		
			2.5%	**5%**	**10%**
2.5%	100.14^a	5.94		-5.20 (.056)	-10.77 (.000*)
5%	105.34^a	7.03	5.20 (.056)		-5.57 (.038*)
10%	110.90^b	7.86	10.77 (.000*)	5.57 (.038*)	

ANOVA analysis and Post-hoc Tukey test were used for comparison of mean flexural strength (MPa) of 2.5 wt %, 5 wt %, and 10 wt % filler reinforced groups.

Mean difference was calculated from [I]-[J]

** p-value less than 0.05 was considered to be statistically significant.*

**The letter "b" superscript meant significant difference between groups (p<0.05).*

Table 4 *The effect of size, percentage by weight, and absence of aluminum oxide particle reinforcement on the flexural strength of heat-polymerized acrylic resin*

Weight % and size of aluminum oxide particle [I]	Mean flexural strength (MPa)	Standard deviation	Mean difference (p-value) [J]						
			Group C	Group NA1	Group NA2	Group NA3	Group A1	Group A2	Group A3
Group C	99.25^a	3.94		-1.75 (-.997)	-7.00 (.255)	-12.10 (.003*)	-0.02 (1.000)	-5.17 (.615)	-11.19 (.008*)
Group NA1	101.00^a	5.72	1.75 (.997)		-5.25 (.596)	-10.36 (.018*)	1.73 (.997)	-3.42 (.917)	-9.45 (.041*)
Group NA2	106.25^a	6.02	7.00 (.255)	5.25 (.596)		-5.10 (.629)	6.99 (.257)	1.84 (.996)	-4.19 (.808)
Group NA3	111.36^b	6.26	12.11 (.003*)	10.36 (.018*)	5.10 (.629)		12.09 (.003*)	6.94 (.265)	0.91 (1.000)
Group A1	99.27^a	6.34	.018 (1.000)	-1.73 (.997)	-6.99 (.257)	-12.09 (.003*)		-5.14 (.619)	-11.18 (.008*)
Group A2	104.42^a	8.13	5.17 (.615)	3.42 (.917)	-1.84 (.996)	-6.94 (.265)	5.15 (.619)		-6.03 (.431)
Group A3	110.45^b	9.50	11.19 (.008*)	9.45 (.041*)	4.19 (.808)	-.9110 (1.000)	11.18 (.008*)	6.03 (.431)	

ANOVA analysis and Post-hoc Tukey test were used for comparison of mean flexural strength (MPa) of 2.5 wt %, 5 wt %, and 10 wt % filler reinforced groups.

** p-value less than 0.05 was considered to be statistically significant.*

**The letter "b" superscript meant significant difference between groups (p<0.05).*

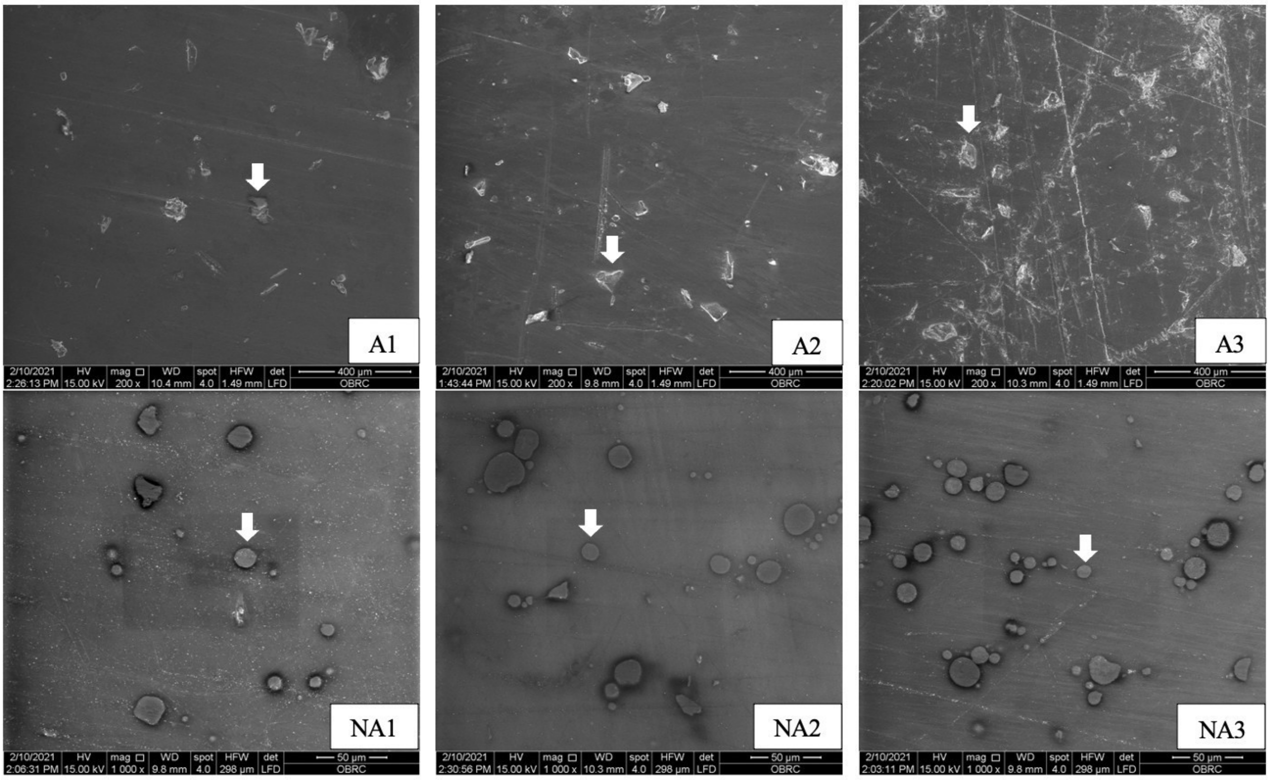

Figure 4 *Scanning electron microscope images of PMMA specimens. The white arrow indicates the reinforced particle. A1: polished surface of 2.5% alumina reinforced PMMA specimen (200x magnification), A2: polished surface of 5% alumina reinforced PMMA specimen (200x magnification), A3: polished surface of 10% alumina reinforced PMMA specimen (200x magnification), NA1: polished surface of 2.5% nano-alumina reinforced PMMA specimen (1000x magnification), NA2: polished surface of 5% nano-alumina reinforced PMMA specimen (1000x magnification), NA3: polished surface of 10% nano-alumina reinforced PMMA specimen (1000x magnification)*

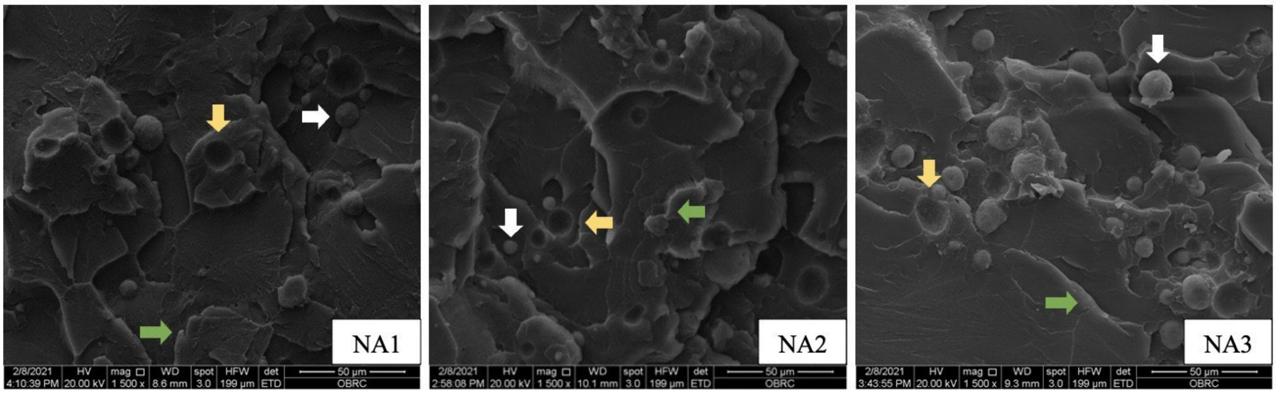

Figure 5 *Scanning electron microscope images of poly-methylmethacrylate (PMMA) specimens. The white arrow indicates the reinforced particle, the yellow arrow indicates the area that reinforced particle diminished and area of adhesive failure, and the green arrow indicates area of cohesive failure. NA1: fractured surface of 2.5% nano-alumina reinforced PMMA specimen (1500x magnification), NA2: fractured surface of 5% nano-alumina reinforced PMMA specimen (1500x magnification), NA3: fractured surface of 10% nano alumina reinforced PMMA specimen (1500x magnification)*

Table 5 *Mode of failure of each group of all specimens*

Group	n (specimens)	Adhesive	Cohesive	Mixed
C	10	0%	100%	0%
A1	10	0%	0%	100%
A2	10	0%	0%	100%
A3	10	0%	0%	100%
NA1	10	0%	0%	100%
NA2	10	0%	0%	100%
NA3	10	0%	0%	100%

Discussion

The present study investigated the effects of 2.5% by weight, 5% by weight, and 10% by weight of silane-treated nano-aluminum oxide (Al_2O_3) particles and aluminum oxide particles reinforcement on the flexural strength of heat-polymerized acrylic resin denture base. A previous study reported that the ultimate flexural strength of a material reflects its potential to resist catastrophic failure under a flexural load [30]. Thus, flexural strength was chosen as the independent factor in this study.

Many studies in the past have shown that the reinforcement of PMMA with various types of fibers and fillers such as polyaramid fibers, carbon fibers, glass fibers, ultra high modulus polyethylene fibers, metal powders, zirconia oxide, and sapphire whiskers, improved its mechanical properties. [4,9,31,32] Ceramic filler is more preferable for reinforcement of acrylic resins than metal filler due to its low density. For instance, the density of sapphire (3.99 g/cm^3) is considerably less than that of Co (8.9) and Cr (7.1), therefore the light weight of acrylic resin denture bases is retained. Furthermore, these ceramic powders have the advantage of being white, so they are less likely to alter the finished appearance of the denture base material than metal powders.[4] Moreover, previous studies demonstrated an increase in flexural strength by adding metal oxides (alumina, tin, silver and zirconia) and sapphire whiskers.[4,9,33] An addition of alumina improves the properties of acrylic resin. For example, an increase in thermal conductivity results from alumina reinforcement on PMMA.[34] Also, reinforcing PMMA with alumina leads to higher flexural strength, impact strength, compressive strength, tensile strength, and surface hardness of the acrylic resin.[4,9,33,35] A previous study reported that current silane coupling agents promote the bonding of resin composite to dental restorative materials.[36] Also, the addition of 3-(trimethoxysilyl) propyl methacrylate in hydroxyapatite (HA) reinforced poly methyl methacrylate (PMMA) could improve the mechanical properties by strengthening the chemical bonding and increasing mechanical interlocking between HA and PMMA.[37] Therefore, the application of silane on nano-alumina and alumina surface prior to being reinforced in heat cure acrylic resins was used in the study.

The result of the study demonstrated that the flexural strength of heat-polymerized acrylic resin significantly increased after the incorporation of 10% by weight of silane-treated nano-alumina and alumina particles ($p<0.05$). A reasonable explanation was that there was proper distribution and bonding of the filler within the matrix. Similarly, Chaijareenont P. *et al* presented that 10% by weight of treated particles with silane coupling agent 18-23 µm particles led to a 23.86 % increase in flexural strength.[25] In addition, 10% and 15% by weight of aluminum fillers which were added in the denture bases caused an increase in flexural strength.[4,21] Although increase in weight % of alumina results in improvement of flexural strength of acrylic, the enhancing of void formation may occur. In the present study, aluminum oxide particles were pretreated with silane, which significantly improved the bond strength of alumina particles to PMMA. Therefore, the chances of void formation were reduced and flexural strength significantly improved in higher weight % reinforced groups, specifically 10% by weight alumina reinforced groups.[29]

Increased flexural strength can also be explained via the phenomenon of transformation. Al_2O_3 is found in many crystalline phases, and all filler particles revert to the most stable hexagonal alpha phase at elevated temperatures. Its structural application is interesting.[4] Transformation phenomenon occurs and reduces crack propagation when enough stress develops and microcracks begin to propagate. As a consequence, proper distribution of the filler within the matrix is able to stop or deflect cracks.[38] SEM examination of specimens reinforced with 2.5%, 5% and 10% by weight of silane- treated nano-aluminum oxide (Al_2O_3) particles and aluminum oxide particles showed a wide, equal distribution of oxide particles within the resin matrix (Fig. 6). Similarly, Vojdani M, *et al* presented that specimens added with 2.5% by weight of aluminum oxide have even distribution from the SEM evaluation. It confirmed that the technique of mixing particles using a magnetic stirrer is acceptable and reliable. A reasonable explanation was that using the magnetic stirrer provided uniform consistency and reduced human errors.[28] In addition, the SEM examination of the fractured surface of specimens revealed that the mode of failure for most specimens was a mixture of cohesive and adhesive failure. Fracture was found between the interface of silane-treated aluminum oxide particles and PMMA (adhesive failure), and between PMMA and PMMA (cohesive failure). It could be explained that aluminum oxide reinforced particles were evenly distributed in resin specimens, so most of the fractured areas were a mixture of cohesive and adhesive failure. There was hardly any adhesive failure.

One of the limitations of this study was that material fatigue by thermocycling in water or artificial saliva with or without reinforcement was not carried out to predict the success of material in a clinical environment, as the study was conducted in an in vitro setting. Moreover, although specimens were preselected using an exclusion criteria to reject specimens with surface voids, PMMA has the tendency to generate submicrometer voids.[39] Therefore, fracture resistance of specimens may be affected by void formations located within the specimens. Another limitation of this study was that the SEM images revealed that nano-sized aluminum oxide particles and micro-sized aluminum oxide particles showed a difference in shape. The nano-aluminum oxide particles were spherical in shape, however the micro-sized aluminum oxide particles were irregular in shape. Thus, the micro-sized aluminum oxide particles were a range whereas the nano-sized aluminum oxide particles were more discrete. The difference in shape of aluminum particles resulted from the preparation method. A controlled manner by means of surface tension changed the shape of aluminum particles from a spattered to spherical one. Surface tension also resulted in alteration of the particle-size distribution.[40] Even though change in color of alumina reinforced specimens was not obvious, it was better to use in invisible denture areas such as lingual flange of lower dentures or the palatal area of upper dentures because discoloration and opacity of PMMA particularly could happen when added at high concentration.[23,41]

Micro-sized alumina particles (50-70 µm) is one of the compositions of sandblast used in a dental field. They can be used to reinforce the denture base in order to improve the flexural strength of the denture base because it is available and inexpensive. Enhancing flexural strength could promote the long-term clinical success of the prosthesis and patient satisfaction. However, further research is required to examine cytotoxicity, aging effect and physical properties such as color and surface roughness of these reinforced denture base materials before clinical application. Also, other mechanical properties of nano-alumina or alumina reinforced acrylic resin such as thermal conductivity and impact strength should be considered.

Conclusion

Based on the mentioned results and the limitations, the following was concluded.

1. Changing the weight % of silane-treated alumina reinforced and influenced the flexural strength of heat-polymerized acrylic resins. Reinforcement with silane-treated nano-alumina and silane-treated alumina on heat-polymerized acrylic resins improved the flexural strength of the acrylic resins only when used in an adequate amount (10% by weight).

2. Modifying the particle size (micro-sized and nano-sized) of silane-treated alumina reinforcement generated an indifferent effect on the flexural strength of heat-polymerized acrylic resins. The effect was an improvement in the flexural strength of the resins.

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