The effect of silanized alumina nano -fillers addition on some physical and mechanical properties of heat cured polymethyl methacrylate denture base material

Ban Saad Jasim, B.D.S., M.Sc. [(1)]
Intisar J. Ismail, B.D.S., M.Sc., Ph.D. [(2)]

ABSTRACT

Background: Polymethyl methacrylate (PMMA) is the most commonly used material in denture fabrication. The material is far from ideal in fulfilling the mechanical requirements. Midline fracture; poor thermal conductivity and water sorption, are common problem in this material. The purpose of this study was to evaluate the effect of addition of surface treated Aluminum oxide nano fillers on some properties of heat cured (PMMA).

Materials and methods: In addition to controlled group of heat cured PMMA the silanized (Al_2O_3) nanoparticles was added to PMMA powder by weight in three different percentages 1wt%, 2wt% and 3wt%, mixed by probe ultra-sonication machine. 200 specimens were constructed and divided into 5 groups according to the test (each group consist of 40 specimens) and each group was subdivided into 4 sub-groups. The tests conducted were thermal conductivity, thermal diffusivity, transverse strength, indentation hardness (shore D), surface roughness, water sorption and solubility. The results were statistically analyzed using ANOVA and Dunntt t-test.

Results: A highly significant increase in transverse strength was observed with the addition of (Al_2O_3) nanoparticles to (PMMA) at the percentage of 1wt%, the value was 117.72 Mpa and significant increase at 2wt%; while a significant reduction occurred in transverse strength at the percentage of 3% the value was 90.110 Mpa. A significant increase in surface hardness and non significant differences in surface roughness were observed for all percentages.

Conclusion: The addition of Al_2O_3 nanoparticles to acrylic resin improves the thermal properties and transverse strength of acrylic resin at the same time this addition decreases water sorption and solubility.

Key words: Silane, alumina nanofillers, transvers strength. (J Bagh Coll Dentistry 2014; 26(2): 18-23).

INTRODUCTION

There have been many materials used for denture base such as polymethylmethacrylate (PMMA) resin, modified PMMA resin and nylon [(1)]. The material most often used to fabricate denture base and denture teeth was PMMA resin which is most commonly used material since 1930 [(2)].

PMMA is the most commonly used material due to its satisfactory mechanical and physical properties, compatibility with oral tissue, aesthetics, ease of repair and low cost [(1)].

However, few but important disadvantages are inherent in this resin such as poor strength particularly under fatigue failure inside the mouth, high coefficient of thermal expansion that causes internal stresses to be released during the processing resulting in dimensional inaccuracy, some problems such as denture fracture and wear of the denture teeth still exist [(3-6)].

Recently, much attention has been directed toward the incorporation inorganic nanoparticles into PMMA to improve its properties. The properties of polymer nanocomposites depend on the type of incorporating nanoparticles, their size and shape, as well as the concentration and interaction with the polymer matrix [(10)].

PMMA has low thermal conductivity approximately 0.2 w/m.c [(7)] compared with gold or cobalt alloy denture base material and this can present problems during denture processing as heat produced cannot escape , leading to a temperature rise and this may lead to porosity during fabrication . From the patient's point of view, the problem with low thermal conductivity is that the denture isolates the oral soft tissues from any sensation of temperature. This can lead to patient consuming hot drink without realizing it, which may lead to the back of the throat and possibly even the esophagus being scalded [(8)]. In order to overcome these problems, several attempts were made to modify and improve the strength, thermal properties, and hardness of the PMMA. These attempts included the addition of filler particles such as zirconia, glass fiber, alumina, tin, and Copper or addition of whisker to resin [(9)].

Nanoparticles were undergone surface treatment with silane coupling agent and embedded into PMMA [(21)]. Alumina nanoparticles were treated with 3-(methacryloyloxy) propyltrimethoxysilane (MPS) to get PMMA/alumina nanocomposite with improved properties over pure PMMA.

(1) M.Sc. Student. Department of Prosthodontics, College of Dentistry, University of Baghdad.
(2) Assistant Professor. Department of Prosthodontics, College of Dentistry, University of Baghdad.

MATERIALS AND METHODS

Transverse strength test

The specimens used were prepared with dimensions of (65mm x 10mm x 2.5mm) according to [11]. Ten specimens for each concentration plus the control will make total of (40) specimens for the measurement of transverse strength. All the specimens were immersed in distilled water on the incubator at 37°C for (48) hours before testing.

Test was performed using a universal Instron testing machine, each specimen was positioned on the bending fixture which consists of two parallel supports (50)mm apart, the full scale was 50 Kg ,and the load was applied with a cross head speed of 1mm/min by a rod placed centrally between the supports making deflection until fracture occurs.

Surface roughness test

Specimens with dimension of (65mm x 10 mm x 2.5 mm) were prepared to be used for surface roughness test. Ten specimens for each group were taken to make total of (40) specimens for the measurement of surface roughness test. All specimens were immersed in distilled water at 37°C for 48 hours before being tested [11]. The profilometer device (surface roughness tester) was used to study the effect of Al_2O_3 Nano fillers reinforcement on the micro geometry of the test surface .

Surface hardness test

The specimens were prepared with dimensions of (65mm x 10mm x 2.5mm) according to the American dental association specification [11]. Ten specimens for each concentration plus the control will make a total of (40) specimen for the testing of surface hardness. All specimens were immersed in distilled water for (48) hours before testing [11]. Test was performed using durometer hardness tester (shore D hardness) according to the American dental association specification [11] which is suitable for acrylic material. The instrument consists of a blunt pointed indenter (0.8 mm in diameter) that present in a cylinder (1.6mm in diameter) .The indenter was attached to a digital scale that is graduated from 0 to 100 unit. The usual method was to press down firmly and quickly on the indenter and recorded the maximum reading as the shore D hardness, measurements were taken directly from the digital scale reading. Five measurements were recorded on different areas of each specimen and an average of these five readings was recorded.

Thermal properties tests

Ten specimens were prepared for each group (Total: 40 specimens). The discs were with dimensions of 40 mm in diameter and 2.5 mm in thickness and according to instrument specifications. The hot disk thermal constant analyzer can be used for measuring thermal transport properties of a large variety of materials with thermal conductivities ranging from 0.005w/m.c (Evacuated powders) to 500 w/m.c (graphite). Naturally the parameter heating power measuring time and radius of disk, by which experiment are controlled and must be selected with care in order to arrive at results within the given limits of accuracy. The hot disk sensor consist of an electrically conducting pattern in the shape of double spiral extend out of a thin sheet of Nickel. The Nickel foil was chosen because of its high and well known temperature coefficient of resistivity. The conducting pattern was supported on both sides with a thin electrically insulating material.

The equipment connected to computer that is programmed for the test. By selection experiment type and setting for that type, the method will be selected automatically. The experiment was called TPS (transient plane source).

Figure 1: Instron testing machine

Figure 2: Thermal conductivity test machine

Water sorption and solubility test

Acrylic disc specimens were prepared by using plastic model with dimension of (50 mm ± 1 mm in diameter and 0.5 mm ± 0.1 mm in thickness) [(11)]. Ten specimens of each concentration make total of (40) specimens for measuring of water sorption and solubility. The specimens were dried in dissecator containing freshly dried silica gel. The discs was stored in an incubator at a 37°C ±2 °C for 24 hours after that the specimens were removed to room temperature for one hour then weighted with a digital balance within accuracy of (0.000lg). This cycle was repeated every day at the same time until a constant mass "conditioned mass" (M1) was reached which mean the weight loss from each disc not more than 0.2mg in 24 hours. The specimens then immersed in distilled water for 7 days at 37 ^{0}C ± 2 ^{0}C , after this period of time, each disc was removed from the water with tweezers and wiped for 30 seconds by clean dry hand towel, left in air for 15 second then weighted ,this value represent M2. In order to obtain the value of solubility, the discs were again reconditioned to a constant mass in the desiccators at 37°C ±2 °C as done in the first time for sorption test and the reconditioned mass was recorded as (M3). The whole group was reached to M3 within 5 days.

RESULTS

Data were translated in to computerized database structure. Statistical analyses were done by using SPSS version 16 (statistical package for social science). The statistical analysis includes the following: Arithmetic mean and standard deviation and error, statistical tables and graphical presentations.

Transverse strength test

Table 1: Transverse strength parameters MPa (N/mm^2)

Sample	Mean	S.D.	S.E.	Min.	Max.
Control	97.30	3.6772	1.1628	91.2	100.8
1wt%	117.	9.2679	2.930	102	130.
2wt%	104.	7.6494	2.418	96.	115.
3wt%	90.1	6.1276	1.937	84.	98.4

From table 1 and figure 3 which plots the different means of transverse strengths across different concentrations of the added Al_2O_3 nanoparticles show that the highest mean value appeared in 1wt% which was **117.72 MPa** contrast to lowest value at 3wt% with mean of **90.110 Mpa.**

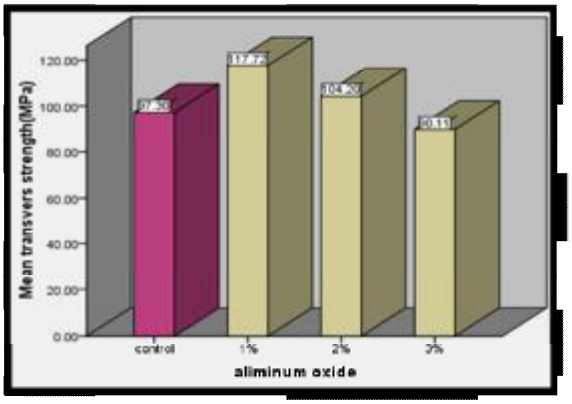

Figure 3: Bar chart of means of transverse strengths

Thermal conductivity

Table 2: Thermal conductivity parameters analysis (w/m.c)

Sample	Mean	S.D.	S.E	Min	Max
Control	.2340	.00776	.002	.22	.24
1wt%	.2398	.00522	.001	.23	.24
2wt%	.2488	.00651	.002	.24	.26
3wt%	.2640	.01308	.004	.24	.27

The highest mean value of thermal conductivity appeared in 3wt% group with a mean of **.2640 (w/m.c)** and the lowest mean was in 0wt% which was **.2340(w/m.c)**. The four groups' means were plotted as bar chart in figure 4.

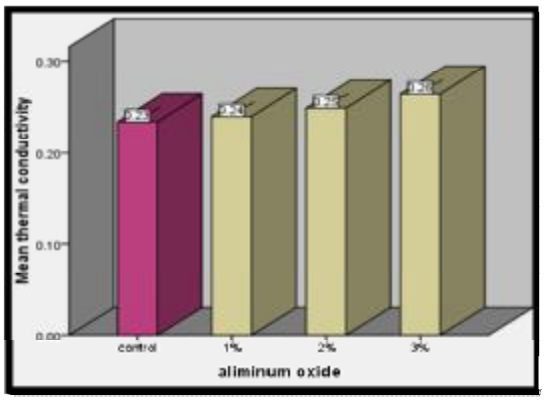

Figure 4: Bar chart of mean thermal conductivity

Thermal diffusivity test

Table 3: Thermal diffusivity parameters analysis (mm^2/sec)

Sample	Mean	S.D.	S.E.	Min	Max
Control	.320	.0596	.0188	.24	.40
1wt%	.336	.0833	.0263	.25	.48
2wt%	.346	.1257	.0397	.03	.45
3wt%	.378	.1199	.0379	.29	.58

The highest mean value appeared in 3wt% with a mean of **.3784 mm^2/sec** and the lowest

mean was in 0 wt% which was **.3206 mm^2/sec.** The four groups' means were plotted as in figure 5.

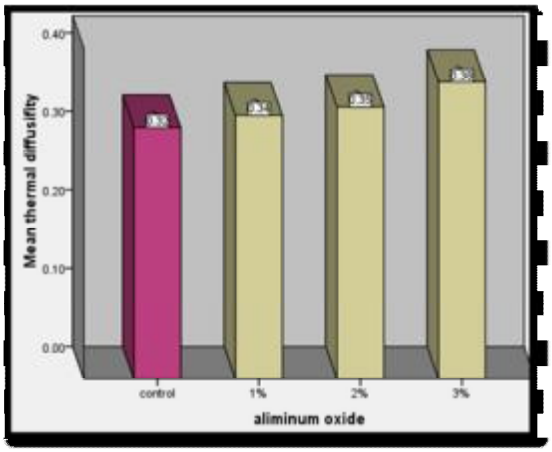

Figure 5: Bar chart of mean thermal diffusivity

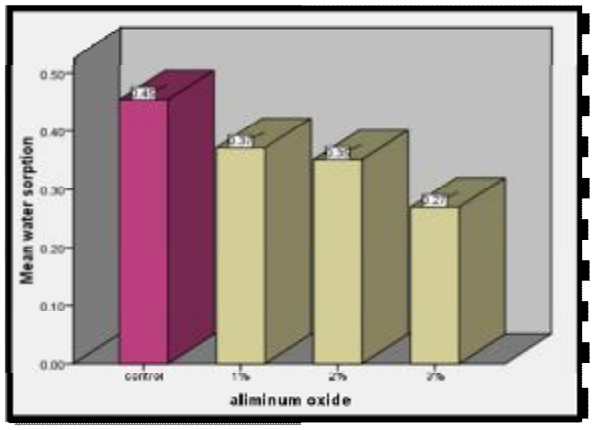

Figure 6: Bar charts of mean value of water sorption

Indentation hardness test

Means, Standard deviations, standard error of mean value, minimum and maximum values of the hardness test of the acrylic resin were listed in table 4 for different groups of different concentrations of the silanized Al_2O_3 nanoparticles that had been added. From table 4 the results of the test indicate that the highest mean value appeared in 3% group with a mean value of indentation resistance **87.3** and the lowest mean value was in 0% (control) which was **85.4**.

Table 4: Hardness parameters analysis

Sample	Mean	S.D.	S.E.	Min.	Max.
Control	85.400	1.26491	.40000	84.00	87.
1wt%	86.000	.66667	.21082	85.00	87.
2wt%	86.500	1.43372	.45338	85.00	89.
3wt%	87.300	.82327	.26034	86.00	88.

Surface roughness test

Table 5: Descriptive data of surface roughness test result (µm)

Sample	Mean	S.D.	S.E.	Min.	Max.
Control	1.2288	.15395	.04868	1.01	1.49
1wt%	1.2289	.20661	.06533	1.01	1.64
2wt%	1.2289	.21664	.06851	.90	1.64
3wt%	1.2290	.29255	.09251	.70	1.60

The highest mean value appeared in 3wt% (mean = **1.229µm**) and the lowest mean was in 0wt% (control group mean = **1.2288 µm**). The four groups' means were plotted**.**

Water sorption test

Table 5: Descriptive data water sorption parameters (mg/cm^2)

Sample	Mean	S.D.	S.E.	Min.	Max.
Control	.4544	.05004	.01582	.36	.50
1wt%	.3713	.05913	.01870	.30	.46
2wt%	.3509	.05613	.01775	.30	.46
3wt%	.2693	.05913	.01870	.20	.36

Water sorption seems to decrease as the concentrations of the added Al_2O_3 nanoparticles was increased, but for a limited extent**.**

DISCUSSION

Transverse strength test

The transverse strength test, one of the mechanical strength tests, is especially useful in comparing denture base materials in which a stress of this type is applied to the denture during mastication [12]**.** The transverse (flexural) strength is a combination of compressive, tensile and shear strengths, all of which directly reflect the stiffness and resistance of a material to fracture [13,14]. The addition of nonsilanized alumina nanoparticles to PMMA revealed lowering the values of transverse strength. This result supported the previous reports that only adding alumina did not improve the mechanical properties of PMMA [6]**.** This could be due to the lack of interfacial bonding between the fillers and resin matrix that deteriorates mechanical properties. Pisaisit et al [5] added silanized and nonsilanized alumina to

PMMA, and by SEM image they found there was a gap between nonsilanized alumina particles and resin matrix this could explain reduction in mechanical properties. The addition of silanized Al_2O_3 nanoparticles in present study increased the values of transverse strength with 1wt% and 2% significantly compared to control groups, then the transverse strength began to decrease with 3wt% in which the value of transverse strength was less than control as in table 1 and figure 3.

Surface hardness test

In this study, shore (D) hardness tester was used which is suitable for measuring the hardness of acrylic resin [15]. Hardness defined as the resistance of material to plastic deformation typically measured under an indentation load .It is a measure of the resistance to wear or scratching [14].

It was found in this study that hardness value showed a non significant increase with 1wt % alumina nanoparticlesr and significant increase with 2wt%, and a highly significant increase with 3wt%.The increase in hardness was directly proportional with the increase in alumina nanoparticles content. This finding is in agreement with previous investigations [16] who concluded that reinforcing dental restorative resins and acrylic resin with ceramic particles (alumina) can produce some improvements in the surface hardness. This increase in hardness may due to inherent characteristics of the Al_2O_3 nanoparticles. Al_2O_3 possesses strong ionic interatomic bonding, giving rise to its desirable material characteristics, that is, hardness and strength. The most stable hexagonal alpha phase Al_2O_3 is the strongest and stiffest of the oxide ceramics. Therefore, it is expected that when Al_2O_3 nanoparticles disperse in a matrix, they increase its hardness and strength.

Surface roughness test

In this study profilometer device was used to estimate the effect of adding silanized alumina nanoparticles on the surface roughness of the specimens. The surface roughness of the acrylic denture base was not significantly change when different percentages of silanized nanoparticles were added. This result may be due to that the alumina nanoparticles have very small size and well dispersion, also surface roughness test is concerned with the outer surface and not the inner surface of the nanocomposite so when small percentage of nanoparticles were added to the acrylic resin only few particles involved on the surface of the specimens . The result of this study coincides with the result of Abdul Ameer [16] when titanium and zinc oxide powder added to the acrylic resin.

Water sorption test and Water solubility test

The absorption of water by acrylic resins is a phenomenon of considerable importance. Acrylic resins absorb water slowly over a period of time, primarily because of the polar properties of the resin molecules. High equilibrium uptake of water can soften an acrylic resin because the absorbed water can act as a plasticizer of acrylic and reduce the strength of the material [17]. The mechanisms which responsible for the water sorption was diffusion which was defined as the migration of one substance through a space or within a second substance in which water will penetrate acrylic resin mass and occupy a position between polymer chains [18]. From the results there were highly significant decreased in the value of water sorption as the percentage of Al_2O_3 nanoparticles increased. The reasons why Al_2O_3 added groups exhibited significantly lower water sorption values than control group can be explained in several ways. During the polymerization process of acrylic resins, porosity or microvoids can occur among polymer chains. A high level of porosity or microvoids has been shown to facilitate fluid transport into and out of polymer by serving as sites for molecules to be sequestered and leading to enhanced solvent uptake and elution. Al_2O_3 nanoparticles used are insoluble in water and could reduce the overall volume of the absorbing polymer. Solubility represents the mass of the soluble materials from the polymers. The soluble materials present in denture base resins are initiators, plasticizers, free monomer and some pigmentation [19]. There were significant decrease in the values of water solubility with the increase in percentage of silanized Al_2O_3 nanoparticles, this decreases could be attributed to the fact that Al_2O_3 is insoluble in water so that the addition of Al_2O_3 to the mass of the specimens would act as additives and their presence will lead to reduction in the solubility of acrylic resin. However, the results were within the limitation of the American dental association specification [11].

Thermal conductivity test

One of the important thermal properties of dental materials are thermal conductivity or ability to transmit heat which obtained from determining the rate at which heat can be transmitted through a given cross sectional area of the specimens of material during a given time interval [20]. Table 2 was showed that there were a highly significant increase in the values of thermal

conductivity with the addition of alumina nanoparticles . This may due to overlapping of thermal conductive nanoparticles inside the polymer matrix to bridge the insulating effect of PMMA matrix. The increase in the amount of fillers make the nanoparticles approximate from each other and increase overlapping of thermal conductive particles that form pathway and permit transition of heat from one side of specimens to another side thus increasing thermal conductivity.

Thermal diffusivity test

The thermal diffusivity describes the rate at which a body with a non uniform temperature approaches equilibrium. As in the thermal conductivity, thickness of the material is important. Thermal diffusivity of acrylic resin was 0.123 mm^2/sec. **(20)**. Table 4 showed that there was an increase in the value of thermal diffusivity with the increase in the percentage of alumina nanoparticles compared to the control group. An overall improvement of thermal diffusivity of PMMA upon the addition the alumina nanoparticles can be attributed to the formation of thermally conducting pathway within polymer matrix.

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