

Evaluation the Effect of Adding Silanized Silicon Dioxide Nano Filler and Carbon Nanotube Composite on Some Properties of Heat Cured Acrylic Denture Base Material

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Abstract:

Background: *The mechanical properties of polymethyl methacrylate (PMMA) are still far from ideal to fulfill the perfect mechanical requirements for dental applications. The purpose of this study was to evaluate the effect of addition of silanized silicon dioxide Nano filler (SiO₂) and carbon nanotube on some properties of heat cured acrylic resin denture base material.*

Materials and methods: *One hundred sixty (160) prepared specimens were divided into 4 groups according to the tests, each group consisted of 40 specimens and these were subdivided into 4 groups (unreinforced heat cured acrylic resin as control group),reinforced acrylic resin with (1%wt carbon nanotube and*

3%wt silanized silicon dioxide) group and reinforced acrylic with(1.5%wt carbon nanotube and 5%wt silanized silicon dioxide)and reinforced acrylic with (1.5%wt carbon nanotube and 7%wt silanized silicon dioxide) .The tensile strength, transverse strength, indentation hardness(shore D), water sorption and solubility were investigated . The results were statistically analyzed using ANOVA test with LSD multiple comparisons.

Results: The result of this study shows a highly significant increase in tensile strength, surface hardness, transverse strength and significant decrease water sorption and water solubility with studied groups (the addition of carbon nanotube and silanized silicon dioxide) to PMMA than control group, also the best results were obtained with acrylic resin reinforced with(1.5%wt carbon nanotube and 5%wt silanized silicon dioxide).

Conclusion: The addition of a mixture of carbon nanotube (1.5%wt) and silanized nano silicon dioxide (5%wt) to PMMA acrylic resins improves its mechanical and physical properties.

Keywords: Carbon nanotube, silicon dioxide, polymethyl methacrylate (PMMA)

Introduction

Polymer used successfully in various application in dentistry for many years Poly methyl methacrylate (PMMA) was introduced as sheet form by Rohm and Hass in 1936 and as powder by Nemours in 1937. Dr. Walter Wright choice PMMA in 1936 for construction denture base for partial and complete denture to restore function,

physiology and esthetic of partial edentulous or complete edentulous patients, because of its properties. [1, 2]

PMMA has several restraints in mechanical characteristics such as polymerization shrinkage, weak flexural and low fatigue resistance and impact strength which leads to the failure of denture. [3-6]. several solutions were developed such as modification of chemical structure or addition of metal wires or plates, fibers, metal inserts. [7-10]

Several experiments note that strengthening and enhancing the PMMA by adding Nano SiO₂ helps improving the impact strength, transverse strength and surface hardness by increasing the concentration of the nanoparticles. [11, 12]

In the field of advanced composites one of the most promising nano – material is Carbon nanotubes because it has unprecedented mechanical, thermal and electrical properties. Carbon nanotubes are strong, adaptable, and lightweight, and generally outline stable round and empty structures. Carbon nanotubes are that single walled and multi walled. [13, 14]

The success of the idea of integrating nanoparticles with nanotubes in order to produce nano composites compounds are characterized by lightness and strength and the cheap price that is the main goal of materials science and engineering applications nowadays. [15] The purpose of this study was to evaluate the effect of addition of silanized silicon dioxide Nano filler (SiO₂) and carbon nanotube on some properties of heat cured acrylic resin denture base material (PMMA).

Materials and Methods

The materials used in this research were Silicon dioxide (SiO₂) Nano filler with Epoxy coupling agent, carbon nanotube, Heat-curing acrylic (powder and liquid), Hard dental stone type III and Separating medium (Tin foil substitutes).

Pilot study was done to select the best concentration of SiO₂ and carbon nanotube. The specimens were divided into 10 groups

according to the concentration of SiO₂ and carbon nanotube as shown in Table 1.

Table 1: Mixing Ratios for all groups.

Group name	Concentration of carbon nanotube by weight	Concentration of SiO ₂ by weight	Amount of polymer (g)	Amount of monomer (ml)
Control	0%	0%	44g	20 ml
A	1%	3%	42.24g	20 ml
B	1%	5%	41.36g	20 ml
C	1%	7%	40.48g	20 ml
D	1.5%	3%	42.02g	20 ml
E	1.5%	5%	41.14g	20 ml
F	1.5%	7%	40.26g	20 ml
G	2%	3%	41.8g	20 ml
H	2%	5%	40.9g	20 ml
J	2%	7%	40.04g	20 ml

For each group transverse strength and hardness test were done and the best results were obtained for group A , E and F . One hundred and sixty specimens were prepared. The specimens were divided into 4 groups according to the tests selected, 10 for each group (control, A, E and F).

Transverse strength test:

Specimens were prepared with dimensions (65mm x 10mm x 2.5mm ± 0.1mm) according to ADA specification no. 12 1999 [11].

Ten specimens for each concentration plus the control will make a total of (40) specimen 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. [11] Test was performed using a universal Instron testing machine (Tinius Olsen, USA). Each specimen was positioned on the bending fixture; 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 hardness testing:

The specimens were prepared with dimensions of (65mm x 10mm x 2.5mm) according to ADA specification no. 12 1999 [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) that was fabricated by (HARTIP 3000 compant) [11] which is suitable for acrylic design material. The instrument consists of a blunt pointed indenter (0.8 mm in diameter) that present in a cylinder (1.6 mm in diameter). The indenter was attached to a digital scale that is graduated from 0 to 100 units. 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 (the same selected area applied for all specimens) and an average of five reading was calculated.

Tensile Strength Test:

Specimens for tensile strength test: Flat dumbbell shape, (16±1mm length, 3±0.2mm width, and 2±0.2mm thickness at the parallel segments). The tensile strength test was conducted according to ISO 527:1993 [16]. An instron testing machine was used to measure the tensile strength of the specimens. All

specimens were placed under tension in a unilateral testing machine at a cross-head speed of 1mm/min. until fracture [17].

Water sorption and solubility test:

Disc specimens were prepared by using plastic pattern having dimensions of (50mm±1mm in diameter and 0.5 mm ±0.1 mm in thickness) according to ADA specification no. 12 1999 [18].

The specimens dried in desiccators containing freshly dried silica gel. The desiccator was stored in an incubator at 37°C ±2°C for 24 hours after that the specimens were removed to a room temperature for one hour then weighted with electronic balance with accuracy of (0.0001g). This cycle of weighting was repeated every day until a constant mass (M1) (conditioned mass) was reached after 7 days, and then all discs of all groups were immersed in distilled water for 7 days at room temperature. The discs were removed from the water with a dental tweezers wiped with a clean dry towel until free from visible moisture, waved in the air for 15 seconds and weighted; this mass was recorded as (M2).

The value of water sorption was calculated for each disc from the following equation:

$$WSP = \frac{M2-M1}{S} \quad (\text{ADA specification No. 12,1999})$$

WSP = Water sorption in mg/cm²

M2 = The mass of the disc after immersion in distilled water (mg)

M1 =The mass of the disc before immersion in distilled water (conditioned mass) (mg).

S = Surface area of the disc (cm²)

In order to obtain the value of water solubility the discs were again reconditioned to a constant mass in the desiccator at 37°C + 2°C as done in the first time for sorption test and the reconditioned mass was recorded as (M3) .

The solubility during immersion was determined for each disc by the following equation:

$$WSL = \frac{M1-M3}{s} \text{ (ADA specification No. 12,1999)}$$

WSL= Water solubility in mg/ cm².

M1= the conditioned mass (mg).

M3= the reconditioned mass (mg).

S= the surface area of the disc (cm²).

Statistical analysis:

The data was analyzed using SPSS Statistical analysis software version 21.0, ANOVA Table with LSD multiple comparison test used to show the significant differences among groups.

Results

The statistical analyses give the following results; first table (2) shows the mean, standard deviation of transverse strength, tensile strength, hardness and water solubility and sorption for the control group and experimental groups.

Transverse strength mean value was much higher when adding mixture of 1.5%wt carbon nanotube and 5%wt silanized silicon dioxide(group E) **31.75 N/mm²** than control **19.25 N/mm²** and other experimental groups.

Tensile strength mean value was much higher when adding mixture of 1.5%wt carbon nanotube and 5%wt silanized silicon dioxide(group E) **0.365 N/mm²** than control **0.161 N/mm²** and other experimental groups.

Hardness mean value was much higher when adding mixture of 1.5%wt carbon nanotube and 5%wt silanized silicon dioxide(group E) **77.31** than control **73.1** and other experimental groups.

Water solubility mean value was much lower when adding mixture of 1.5%wt carbon nanotube and 5%wt silanized silicon dioxide(group E) **0.0118 mg/cm²** than control **0.0863 mg/cm²** and other experimental groups

Water sorption mean value was much lower when adding mixture of 1.5%wt carbon nanotube and 5%wt silanized silicon dioxide(group E) **0.0226** mg/cm² than control **0.1611** mg/cm² and other experimental groups

Table 3 is showing the ANOVA test between control and experimental groups which was statistically highly significant in all tests (transverse strength ,tensile strength, hardness and water solubility and sorption) and table 4 show LSD results for studied groups.

Discussion

The mechanical properties of PMMA were improved by adding different nanofiller , fiber and nanotube , In this study the effect of adding composite material of silanized SiO₂ nanofiller and carbon nanotube on some properties of heat cured acrylic resin denture base material (PMMA) were investigated.

Transverse strength

In most studies transverse strength assess to be as similar to denture prosthesis in the oral environment receiving different masticatory loading force.

In table 2 and 3; the transverse strength in all experimental groups were higher than control group which was statistically significantly improved, this result was in agreement with Fatihallah [11] who found that the addition of silicon dioxide Nano filler (SiO₂) to PMMA improved the mechanical properties of acrylic resin, also agree with Zhou [20] who added carbon nanotube to poly methyl methacrylate denture base causing improvement in transverse strength .

The best result obtained by adding a mixture of 1.5%wt carbon nanotube and 5%wt silanized silicon dioxide which can be related to the very fine size of filler and nanotube which enter between linear macromolecular chains and fill space present and increase strength and rigidity of poly methyl methacrylate denture

base or due to transfer of stress to stiffer and more rigid polymer instead of flexible polymer so depletes energy need to crack propagation .

Surface Hardness test

In Table 2, 3 and 4 an increase in surface hardness of experimental groups appear to be more prominent when compared with control group which was statistically highly significant, this may be due to the action of silicon dioxide as added to carbon nanotube cause decrease hardness of heat cured acrylic resin denture base material (PMMA) because of its elastic property which disagree with Ibrahim [21]. who found that reinforced heat cured acrylic resin denture base material with carbon nanotube significantly decreased hardness.

The increase in hardness in group E (mixture of 1.5%wt carbon nanotube and 5%wt silanized silicon dioxide) can be explained by distributed hard silicon dioxide particle randomly in acrylic denture base materials. Which agree with Fatihallah [11], also slight increase in other experimental groups might be dominated by cross linking density .

Tensile strength

Tensile strength is maximum stress applied to a material which can resistance before fail by localized accelerated deformation [22]. In this study, significant increase in tensile strength was shown in table 2 and 3 when experimental groups were compared with control group. This may be due to either small size of both silanized silicon dioxide and carbon nanotube or due to the effect of coupling agent.

The well distributed and small size nanofiller and nanotube improved modulus and strength and also improve ductility, or might be due to the presence of silicon dioxide and carbon nanotube transfer stress from polymer to nano-particle and nanotube which promoted familiarity between those nanoparticles and polymer and improvement interfacial bounding energy of matrix.

The coupling agent presents more than one active side so lead to molecular interaction by vanderwaal force which enhanced bond strength. These results agree with Hong et al [22] who found the tensile strength increased when modified silica was added to PMMA

Water sorption and solubility

One of the physical properties of acrylic resins is water sorption and solubility, which causes dimensional instability, thereby subjecting the material to internal stresses that may result in crack formation and, eventually, fractures of the denture [23]. From the results seen in table 2 and 3, a highly significant decrease in water sorption in studied groups than the control group which could explained silane coupling agent change polymer action to water from to hydrophilic to hydrophobic, so lead to decrease water sorption.

Solubility is also an important property because it represents the mass of soluble materials from the polymers [24]. water solubility as shown in table 2 and 4 was highly significantly decreased in studied groups than control group which might due to silicon dioxide nanofiller is insoluble in water so decrease water solubility of acrylic resin.

Conclusion

The addition of a mixture of carbon nanotube (1.5%wt) and silanized nano silicon dioxide (5%wt) to PMMA acrylic resins improves its mechanical and physical properties

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Table 2: Transverse strength, tensile strength, hardness and water solubility and sorption mean and standard deviation for the control group and experimental groups

Groups	Control		A		E		F	
Test	mean	S.D.	mean	S.D.	mean	S.D.	mean	S.D.
Transverse strength	19.25	3.63	26.75	3.26	31.75	1.47	21	1.87
Tensile strength	0.1610	0.037	0.236	0.024	0.365	0.047	0.241	0.064
Hardness	73.1	2.400	73.34	1.422	77.31	1.366	74.35	1.677
Water solubility	0.0863	0.0271	0.0780	0.0206	0.0118	0.0042	0.0608	0.0132
Water sorption	0.1611	0.037	0.1191	0.035	0.0226	0.021	0.0732	0.008

Table 3: Transverse strength, tensile strength, hardness and water solubility and sorption ANOVA test between control group and experimental groups

Test	df	Mean Square	F test	Sig.	
Transverse strength	3	162.16	21.942	0.00	HS
Tensile strength	3	0.029	13.863	0.00	HS
Hardness	3	14.981	4.806	0.02	HS
Water solubility	3	162.16	21.942	0.00	HS
Water sorption	3	0.014	18.089	0.00	HS

Table 4: Transverse strength, tensile strength, hardness and water solubility and sorption LSD multiple comparison test between the control group and experimental groups

LSD		Transverse Strength		Tensile Strength		Hardness		Water Sorption		Water Solubility	
		Sig.		Sig.		Sig.		Sig.			Sig.
control	A	.537	NS	.038	S	0.851	NS	0.056	NS	.537	NS
	E	.000	HS	.000	HS	0.006	HS	0.000	HS	.000	HS
	F	.074	NS	.028	S	0.336	NS	0.001	HS	.074	NS
A	E	.000	HS	.002	HS	0.008	HS	0.000	HS	.000	HS
	F	.212	NS	.867	NS	0.434	NS	0.039	S	.212	NS
E	F	.003	HS	.002	HS	0.035	HS	0.025	S	.003	HS

تقييم تأثير اضافة خليط اوكسيد السيلكون النانوي والكربون نانو تيوب على مادة الطقم الاكريليكية الحرارية

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المستخلص

تحسين صفات مادة الطقم الاكريليكية الحرارية هي من اهم مايشغل العلماء في الوقت الحاضر تم استخدمت في البحث 160 عينه قسمت الى اربع مجاميع حسب نوع الفحص احتوت كل مجموعة على 40 وتم فحص بعض الخواص الميكانيكية والفيزيائية واتضح ان خليط الكربون نانو تيوب بنسبة 1.5% من الوزن مضاف اليه اوكسيد السيليكون النانوي بنسبة 5% من الوزن يحسن صفات مادة الطقم الاكريلي الحرارية. الكلمات الرئيسية: اوكسيد السيليكون النانوي، كربون نانوتيوب، الاكريليك الحراري.