Influence of Artificial Weathering on Some Properties of Nano Silicon Dioxide Incorporated Into Maxillofacial Silicone

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Abstract: ***Background**: In spite of desirable properties of Siliconeelastomers like, easy manipulation, easy coloration, andbiocompatibility, it may suffer from degradation after 4-14 months of clinical use. The purpose of this study was to evaluate the effect of addition of silanated SiO_2 nanofiller on the tear strength, tensile strength, hardness and surface roughness of A-2186 maxillofacial silicone after artificial accelerated weathering. **Material and method**: 120 samples were fabricated by addition of 5% SiO_2silanated nanofiller to A-2186 silicone. Based on our pilot study, the 5% reinforcement revealed superior properties (tear strength, tensile strength, and hardness) among the other percentages (3%,4%). The main study samples were divided into four (4) groups, each group contains (30) samples according to the conducted tests, i.e.tear strength, tensile strength, hardness test, and surface roughness.Then, each group was subdivided into three subgroups according to the artificialweathering environment (A.control before weathering, B. after 200 hours of artificial weathering, and C. after 300 hours of weathering), (n= 10 samples in each subgroups.The samples were tested before and after artificial weathering, andthe data were analyzed with a descriptive statistical analysis, one-way ANOVA and post-hoc LSD test. **Results**: The addition of 5%of SiO_2nanofillerinto A-2186 maxillofacial elastomer resulted in a highly increase in mean values of the tear strength, tensile strength and hardness according to the results of our pilot study. After artificial weathering for(200-300 hours), the tear strength and tensile strength values werehighly significantly decreased ($P\leq 0.01$), while the hardness value was highly significantly increased($P\leq 0.01$) with aging but still within acceptable clinical limit. A non-significant decrease in surface roughness($P>0.05$) was observed after artificial aging. **Conclusion**: Incorporation of SiO_2 nanofiller did not protect the silicone matrix from artificial aging degradation, but can increase the service life of the prosthesis.***

Keywords: Maxillofacial prosthesis, silicone elastomer, artificial weathering, nanofiller

1. Introduction

High rates of cranio-maxillofacial injuries cases occurred by explosive devices that caused by terrorist attacks, As a result of these attacks both the civilian and military people will be in danger of maxillofacial trauma and should be prepared for rehabilitation [1]. Usually the plastic surgery was introduced as the first option of treatment but sometimes the surgery choice excluded due to unfavorable condition, rehabilitation with maxillofacial prosthesis is the best choice for improving patient esthetic andconfidence and facilitating their return to society [2,3].Silicone elastomer usually used as best choice in formation of maxillofacial prosthesis due to biocompatibility, easy manipulation, and realistic appearance [4].

Most appropriate maxillofacial prosthesis should have optimum physical and mechanical properties and keeping these properties during service lifetime of prosthesis with high tear strength, tensile strength, and adequate hardness level near the hardness of defect site [5]. Actually the silicone elastomers alone have non adequate physical and mechanical properties with unsatisfactory strength for most cases without adding filler particle, so that adding of filler is a popular technique resulting in increased silicone modules strength and hardness of final product with enhance mechanical properties[6, 7].Zayed et al,2014[8] concluded that the addition of 3% of silanated SiO_2 nanofiller to A-2186 maxillofacial silicone resulted in improvement in the mechanical properties, particularly the tear strength.Cevik and Eraslan,2016[9] assessed the effect of different nanoparticles i.e. titaniumdioxide, fumed silica and silaned silica on the mechanical properties of A-2000 and A-2006 silicone elastomer, the results revealed that A-2000 silicone exhibit superior mechanical strength in all study groups. Other problem with maxillofacial prosthesis is the short service life(4-14 months) with loss of elasticity and degradation that resulted from environmental factors i.e., temperature alteration, UV radiation and daily usage and cleaning of the prosthesis by the patient[2, 10]. Wang etal,2014[11] investigated the effect of TiO_2on the mechanical and antiaging properties of MDX-4-4210 medical silicone, they found a non-significant effect of thermal and UV aging on the tensile strength of 6% reinforced silicone. Nano-sized SiO_2 have " a large surface area with small size particle, active function, and strong interfacial interaction with organic polymer". [6, 8]

Therefore, the present study aims to evaluate the effect of the artificial accelerated weathering on some mechanical properties of RTV (A2186) maxillofacial silicone afteraddition of 5% SiO_2 nanofiller.

2. Material and Methods

At the beginning, a pilot study was conducted to determine the proper percentage of nanofiller to be added to A-2186 maxillofacial silicone. 36 samples were prepared and divided into four groups 0% (control group), 3%, 4%, and 5% silicone reinforced with SiO_2 nanofiller. The tear strength, tensile strength, and hardness properties were tested, the results reaveled that 5% SiO_2reinforcement had greater value among the other percentages. The main study involved 120 samples, they were fabricated from A-2186 maxillofacial silicone (Factor II Inc. USA) after addition of 5%silanated SiO_2 nanofiller (MK Nano Canada). The nanofiller was addedto part A(base) by weight before adding the catalyst,and mixed by vacuum mixer for10 min(first 3 min. without vacuum to avoid suction of nanofiller, then vacuum turned on for remaining 7 minutes. Then part B (catalyst) was weighed and added into the modified basewith a final mixing ratio of 10:1 and mixed by vacuum mixer for 5 minutes.[12]

The resultinghumongous air bubbles free mixture was produced, andthen poured directly into special design plastic custom made mold consist of following parts(Bottom of the mold with frame, cover, 4 G-clamped and four screw, washers, nuts) as shown in fig (1-1). This design of the plastic mold allow the air bubbles to runaway toward the border and produced air bubbles free silicone sheet after complete setting time for 24 hoursat 23°C ±1 according to manufacturer's instructions.

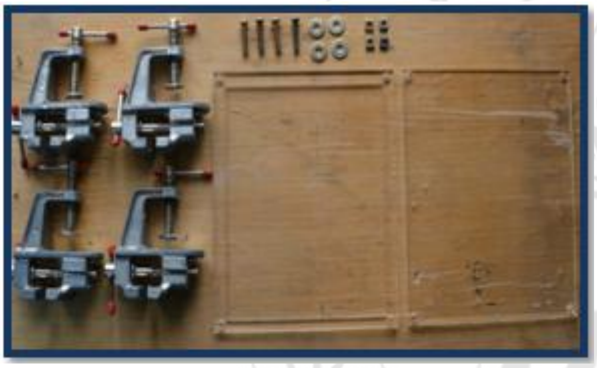

Figure1: Custom made plastic mold parts.

Furthermore the silicone sheet cutting was made by custom made cutter, fabricated according to the ISO 23529:2010[13] for each conducted tests (tear strength, tensile strength,hardness, surface roughness) as shown in fig (2)

Figure 2: Custom made cutters.

The cutting process was made with the aid of hydraulic press whichapplied controlled pressure (MEGE 2012 Spain). After cutting process the samples were labeled and stored in a light proof container at room temperature andtested within 16 hours to avoid changing in physical properties [14]. Then the samples were divided into four groups (4) according to each conducted tests (tear strength, tensile strength, hardness, and surface roughness), with (30) specimens for each group. The testing procedures were performed according to the following:

a) Tear strength test: The specimens were formed and tested according to ISO 34-1:2010 specification [15], angle test samples without neck having a thickness of2±0.2mm. The tear strength was measured by an Instron testing machine (Laryee Technology Co., Ltd., China), the sample was stretched at (500mm/min) until it break, and the maximum force of breakage was recorded.Theresults of tear test were calculated according to the following equation:
   Tear strength=F/d
   Where F, d denote the maximum force (N), and the thickness of sample (mm), respectively.
b) Tensile strength test: The specimens were prepared and tested according to ISO 37:2011 standardization[16]. Type 2 dumbbells-shaped, with thickness 2±0.2mm. The testing was done with the aid ofcomputer controlled testing machine (Instron).The tensile specimens were subjected to stress rate of (500mm/min) until it breaks,and the tensile strength was measured according to the following formula: tensile strength=F/w×twhere F, w, and t represent the maximum force at breakage (N), the width of narrow portion of sample (mm), and the thickness of narrow portion of samples (mm), respectively.
c) Hardness ShoreA test:The specimens were prepared and tested according to ISO 7619-1:2010 specification[17]. Thesample made for hardness test should have a thickness of 6mm at least and the outer outline of sample should accept five point of reading at least 6mm between each other. The dimension of sample fabricated in this study was (25mm×25mm×6mm).
d) Surface roughness:The specimens were fabricated with dimensions (25mm×25mm×6mm) as the same dimensions of hardness sample. Profilometer tester was used in this test, its stylus moves on the surface of the sample, and three(3) measurements were obtained fromeach sample and the average of these reading considered the roughness of the surface.

Furthermore the specimens were subdivided into three subgroups (A,B, and C) according to artificial weathering environment(A-before aging, B-after 200 hours of agingand C- after 300 hours of aging) (10 specimens for each subgroup).

The specimens were assessed in Weathr-Ometer device (QUV) according to the most popular aging standardization ASTM G-154under cycle 7[18]. This cycle is primarily exposing the specimen to 25 cycles of(8) hours of UVA light source at 340nm with high temperature of 60°C, then followed by 4 hours(spray for 0:15minutes) and condensation (dew) at 50°C for 3:45minutes.Two specimens were tested by FTIR spectrometer(8400 Shimadzu, Japan), one of specimen without nanofiller and the other with 5% SiO_2 nanofiller. Furthermore two specimens were tested by Scanning Electron Microscope(SEM;Inspect S50, FEI, USA), one specimen before aging and the other one after 300 hours of artificial weathering.

The data of study were statistically analyzed by descriptive statistics, one way ANOVA, and post-hoc LSD test, with significance level at $P \leq 0.05$, highly significant level at $P \leq 0.01$, and a non-significant level at $P > 0.05$.

3. Results

FTIR, SEM examinations:

FTIR analysis of the specimens before and after addition of SiO_2 nanofiller are shown in fig (3). The change in the peaks within spectra range from3200-3600cm^{-1} due to hydrogen bond formation between nano-silica and silicone matrix. In addition, a change in the transmittance percentage from 60% to 45% was observed within the same range from 3200-3600 cm^{-1}.

Figure 4; show the SEM images before and after artificial aging. The SEM examination revealed a well dispersion of SiO_2 nanofiller within silicone matrix without re-agglomeration of nanoparticles as a result of aging.

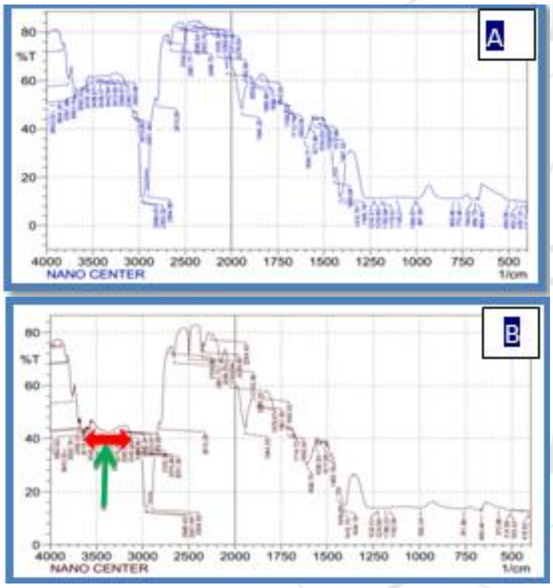

Figure 3: FTIR spectrum of maxillofacial silicone A. before and B. after addition of nano SiO_2.

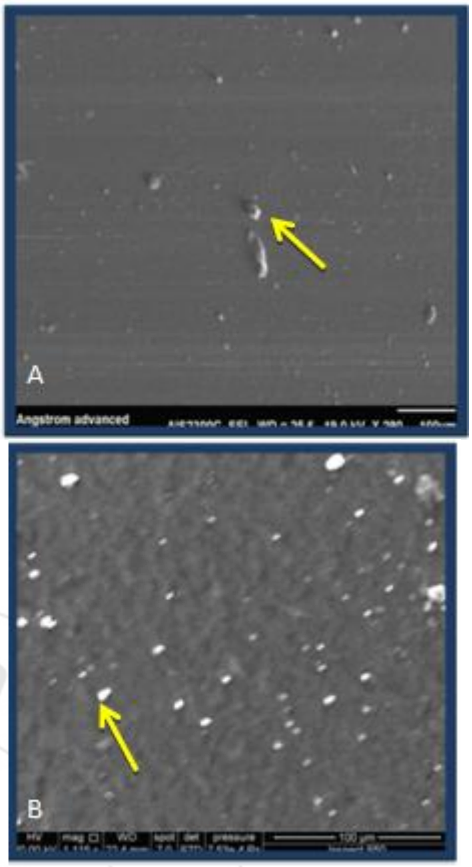

Figure 4: SEM images A. before aging and B. after 300 hours of aging (Arrow: nanoparticle).

Tear strength, tensile strength, surface hardness, and surface roughness:

The means, standard deviation, minimum and maximum values of aforementioned properties were presented in tables 1-4.

The tear strength values were significantly decreased after 200-300 hours of artificial weathering ($p<0.05$) as shown in table 1.

The tensile strength values, also significantly decreasesafter 300hours of artificial weathering ($p<0.05$), while a non-significant decrease recorded after 200 hours of artificial weathering ($p>0.05$) as displayed in table 2.

In contrast to the above parameters, the results of shore A hardness were significantly increased after 200-300 hours of artificial accelerated weathering ($p<0.05$) as shown in table 3. The addition of 5% SiO_2 nanofiller to A-2186 silicone led to a non-significant decrease in surface roughness after 200-300 hours of artificial weathering ($p>0.05$) as presented in table 4.

Table 1: Descriptive statistics, One-way ANOVA and LSD of Tear Strength

	Control A	*After 200h B*	*After 300h C*	*ANOVA F-test*	*Sig.*	*Groups*		*P-value*	*Sig.*
N	10	10	10	7.536	0.003 HS	A	B	0.025	S
Mean	24.49	22.05	20.55				C	0.001	HS
SD	2.51632	2.05968	2.28135			B	C	0.154	NS
SE	0.79573	0.65133	0.72143						
Min.	21.59	18.71	17.61						
Max	28.52	25.15	24.18						

Table 2: Descriptive statistics, One-way ANOVA and LSD of Tensile Strength

	Control A	*After 200h B*	*After 300h C*	*ANOVA F-test*	*Sig.*	*Groups*		*P-value*	*Sig.*
N	10	10	10	4.983	0.014 HS	A	B	0.471	NS
Mean	5.08	4.92	4.42				C	0.005	S
SD	0.49556	0.45464	0.51573			B	C	0.030	HS
SE	0.15671	0.14377	0.16307						
Min.	4.52	3.99	3.48						
Max	5.89	5.77	4.97						

Table 3: Descriptive statistics, One-way ANOVA and Post-hoc test of Shore A Hardness

	Control A	*After 200h B*	*After 300h C*	*ANOVA F-test*	*Sig.*	*Groups*		*P-value*	*Sig.*
N	10	10	10	17.478	0.000 HS	A	B	0.002	HS
Mean	38.47	40.60	42.02				C	0.000	HS
SD	1.61650	1.11305	1.28012			B	C	0.026	HS
SE	0.51118	0.35198	0.40481						
Min.	36.48	38.78	40.77						
Max	41.38	42.29	44.31						

Table 4: Descriptive statistics and One-way ANOVA of Surface roughness

	Control A	*After 200h B*	*After 300h C*	*ANOVA F-test*	*Sig.*
N	10	10	10	1.196	0.318 NS
Mean	0.285	0.243	0.239		
SD	0.08822	0.06236	0.06850		
SE	0.02790	0.01972	0.2166		
Min.	0.183	0.182	0.171		
Max	0.443	0.357	0.358		

4. Discussion

Impairment in mechanical and anti-aging properties was significantly affected in shortening the service life of maxillofacial silicone prosthesis [11]. The service life of the maxillofacial prosthesis also depends on optimum mechanical properties of the material and how the patient used their prosthesis [19].In the present study the specimens were exposed to 300 hours of artificial weathering,equivalent nearly 282.9 days, and this means 9.43 months in outdoor weathering according to Baghdad city [20,21].

The reduction in tear strength values after 200-300 hours of artificial weathering could be due to main changes in polymer structure and modification in polymer network as function of photo-oxidation reaction resulted in molecular weight distribution due to chain scission, continuous cross linking, and production of volatile degradation products.Besidesthat, the effect of nanoparticle addition resulted in further post polymerization cross linking as function of high irradiation energy causing reduces in the strength value of elastomer [22.23].

The results of this study coincide with Hatamleh et al, Nguyen et al and Zardawi et al [24-26]; they reported decrement in tear strength after aging.

While the results disagreed with Nobrega et al 2016[27] who studied the effect of artificial accelerated weathering on (Hardness, tear strength, and permanent deformation) of MDX4-4210 maxillofacial silicone filled with different nanofiller (ZnO, $BaSO_4$, and TiO_2). They found the tear strength increased with artificial aging that could be due to different type of maxillofacial silicone and different type of nanofiller.

The significant decreased in tensile strength values after 300 hours of artificial weathering may be attributed to thesilicone when exposed to high energy of irradiation undergo cross linking, this cross linking was too high, because of high radiation dose that causes degradation in physical and mechanical properties of silicone and resulted in brittle material which can be easily deformed with lower forces [28].

These results in accord withNguyen et al and Eleni et al [25, 29], they found significant decreased in tensile strength after aging. In contrast to the present study Wang et al[11] reported a non – significant change in tensile strength of MDX-4 silicone after thermal and UV artificial aging; this could be attributed to the differences in the nanofiller, silicone material and aging cycle.

The hardness values were significantly increased after 200-300 hours of artificial weathering but still within clinically applicable range between (16-45) shore A depending on the missing facial part[11, 30]. This increase in hardness may be due to continuous polymerization as function of the aging process (UV exposure), along withthe evaporation of polymer ingredient [31, 32]. More over the addition of nanofiller to polymer resulting in filler-filler networks within polymer matrix causes the mechanical interlocking and the inter –matrix spaces decreased so that the material became denser and stiffer with high resistance to penetration.[33]This result wasin agreement withGuiotti et al and Nobrega et al[27, 34]who found increase in hardness after artificial aging.

While the result was in disagreement with Nguyen et al 2013 who studied the effect of artificial accelerated weathering on mechanical properties of maxillofacial silicone (MDX4 - 4210) after adding opacifier and UV absorber, the silicone became softer after artificial weathering, this could be due to different technique used in setting process, also different type of opacifier were added.

The surface roughness values were non-significantly decreased after 200-300 hours of artificial weathering, this might be due to continuous polymerization which promotes further arrangement and supplement of polymer chain leading to fine, smooth silicone surface with the time as function of the aging process [35],also the effect of water spray through the cycle of artificial aging may reduce the surface roughness by water adsorption to the surface of polymer causes swelling of surface by stretching resin matrix [36].

Further studies are suggested to investigatethe addition of SiO_2 nanofiller to anew generation of maxillofacial silicone with additional hours of artificial weathering.

5. Conclusions

Within the limitation of this in-vitro study, it can be concluded that:

1) The addition of SiO_2nanofiller did not protect the silicone elastomer from degradation under aging condition.
2) Both 200 hours and 300 hours of artificial accelerated weathering resulted in reduction of the tensile and tear strength values, while the hardness values were significantly increases but still within acceptable clinical limit. A non-significant decrement in surface roughness of silicone samples was recorded.

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