

Evaluation of Some Properties of Heat Cure Acrylic Resin after the Addition of Salinated Aluminum Silicate Composite Filler

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Abstract: *Background:* improvement of mechanical properties of heat cure acrylic resin with inorganic filler had great attention. The purpose of this study was to evaluate some mechanical properties of heat cure acrylic after the addition of salinated aluminum silicate Al_2SiO_5 filler in different concentrations. *Materials and methods:* A total 124 specimens were constructed, 30 specimens were prepared from heat cure PMMA without additives (control) and 90 specimens were prepared from heat cure acrylic resin with the addition of salinated Al_2SiO_5 composite filler which divided into 3 test groups according to different percentage (3%, 5%, 7%). Each group was divided into 10 sub-groups according to the test performed which was impact strength, surface hardness and surface roughness. Also for each group, 4 specimens were prepared for FTIR test. *Results:* The results of addition salinated Al_2SiO_5 particles to PMMA record a highly significant increase in impact strength, surface hardness and surface roughness. For impact strength test, the highest mean value was for specimens containing 3% Al_2SiO_5 . While for surface hardness test and surface roughness test, there is a marked increase in the mean values as the concentration of fillers increase. *Conclusion:* Within the limitation of this study, it may be concluded that the addition of 3% Aluminum silicate to heat cure acrylic denture base material improves the impact strength, hardness with slight increases the surface roughness.

Keywords: aluminum silicate, fillers, heat cure, strength, roughness

1. Introduction

Poly methyl methacrylate (PMMA) acrylic resin material is preferably used for prosthodontic applications this mainly for its adequate aesthetics and desirable characteristics. It has adequate strength, low water sorption, low solubility and low thermal conductivity and it is free from toxicity⁽¹⁾

However, the mechanical strength of acrylic resin is not sufficient to maintain the longevity of dentures, the main problems associated with PMMA as denture base material are poor strength and fracture due to imbalance occlusion inside the mouth, impact failure outside the mouth.^(2,3)

In denture base, there are weak areas susceptible to fracture such as the midline of the palate, small anterior area of the mandibular denture after severe resorption and high buccal and lingual frenal attachments that under gone fracture⁽⁴⁾ this problem lead to several attempts has been conducted in order to avoid denture fractures, such as increase the denture thickness. The Co-Cr mesh reinforcement produced the highest transverse strength and the metal wire reinforcement produced the greatest increasing in impact strength and tensile strength.⁽⁵⁾

The use of flexible type of denture base materials such as polyamide (nylon) material which is known as a flexible acrylic demonstrate a great impact strength, toughness, and resistance to fracture⁽⁶⁾

Addition of a polyfunctional crosslinking agent to acrylic denture base material such as polyethylene glycol dimethacrylate⁽⁷⁾ or by incorporating a rubber phase⁽⁸⁾ or

fibers⁽⁹⁾ in order to enhance the mechanical properties of denture base materials.

Attention has been directed toward the incorporation of inorganic fillers into acrylic resin to improve its properties. It was noted that reinforcement of PMMA with 2.5% of Al_2O_3 significantly increases the flexural strength and surface hardness of the resin while the surface roughness not differ from the control group⁽¹⁰⁾

Modification of heat-cured acrylic resins with certain amounts of metal oxides done by Neset et al in 2013 who added Al_2O_3 , TiO_2 and ZrO_2 fillers in 1% and 2% by volume for each filler type resulted in significant increase in impact strength and fracture toughness and significant decrease in water sorption and solubility⁽¹¹⁾

The problem with reinforcement of acrylic denture base with fillers is the adhesion failure⁽¹²⁾ so surface modification of an inorganic particle with an organic substance is a useful way to reduce its surface energy and increase its compatibility with polymer matrix and dispersion homogeneity and thus improve the properties of the polymer/ inorganic particles.^(13, 14)

Inorganic filler aluminum silicate that used in this study has been found to be biocompatible material⁽¹⁵⁾, and can be simply processed with acrylic denture base material.

The aims of this study was to investigate the effect of addition aluminum silicate composite fillers into heat cure resin on some mechanical properties including: impact strength, hardness, and surface roughness.

2. Materials & Methods

2.1 Surface modification of aluminosilicate (Al₂SiO₅R) with 3-methacryloylpropyl trimethoxysilane (MPS)

One hundred milliliter of ethanol aqueous solution (70 vol%) was prepared from methanol (*Carloerba, Rodano, Milano, Italy*) 99.8 vol% and deionized water. The PH was adjusted to 4.5 by titrating with acetic acid (*Carlo erba, Milan, Italy*) 99.9% by using a pH meter (*ORION 420A, Orion Research Inc., Boston, MA, USA*). Then, 1.5 g of MPS was added respectively into the ethanol aqueous solution, and stirred. This MPS solution was stored in a 100 mL polyethylene cup with a cover, and allowed 5 min for hydrolysis and silanol formation. Then 30 g of aluminum silicate powder (*The British Drug Houses LTD.B.D.H. Laberayory Chemical Group Poole England*) were added into MPS solution. The mixture was stirred until the solution was completely evaporated, and left dried at room temperature for 14 days.⁽¹⁶⁾

The (FTIR) spectrophotometer (*Shimadzu, FTIR-8400S Japan*) was used to determine whether functional group of the MPS have been attached to fillers or not. This was appeared by analyzing characteristic vibrations of functional groups.⁽¹⁷⁾

2.2 Specimens' preparation

Two different metal patterns were constructed by cutting metal copper alloy plate in desired shape and dimension. For impact strength test, a bar shaped specimen (80X10X4) mm length, width, and thickness respectively.⁽¹⁸⁾ For hardness test, and surface roughness test: a bar shaped specimen (65X 10X 2.5)mm length, width, thickness respectively.⁽¹⁹⁾

Mold for specimens used in this study was following the conventional flasking technique for complete denture.

2.3 Proportioning and mixing of acrylic

Amounts of polymer, monomer and silanized alumina silicate with percentages 3%, 5% & 7% used in the study where weighted by using an electronic balance with accuracy of (0.0001g). The proportioning of acrylic powder/ liquid ratio and the percentage of added filler was revealed in the table 1.

Table 1 The proportioning of acrylic powder/ liquid ratio and the percentage of added filler

Al ₂ SiO ₅ percentage	Amount of Al ₂ SiO ₅	Amount of PMMA	Amount of monomer
0% control	0	12g	6ml
3% test	0.360g	11.640g	6ml
5% test	0.600g	11.400g	6ml
7% test	0.840g	11.160g	6ml

At this step filler mixed with acrylic powder according to Abdulkareem in 2016,⁽²⁰⁾ Then the mixture of acrylic powder/ liquid was done and left to stand until a dough stage was reached. Then packed, flasked, and processed as conventional way for complete denture construction.

The specimens were grouped into one control group and 3 test groups. As shown in Figure 1. Ten specimens for each group make a total of 40 specimens for each mechanical testing.



Figure 1: Acrylic specimens A. control specimen 0% fillers B. acrylic specimen 3% Al₂SiO₅ C. Acrylic specimen 5% Al₂SiO₅ D. acrylic specimen 7% Al₂SiO₅

2.4 Mechanical properties tests

In this research, Mechanical test includes the following:-

1-Impact strength test: this was conducted following the procedure given by the ISO 179 with charpy type impact testing instrument (**Impact tester N. 43-1, INC. USA.**). The specimen was supported horizontally at its ends and struck by a free swinging pendulum which released from a fixed height in the middle. A pendulum of 2 joules testing capacity was used. The charpy impact strength of not notched specimen was calculated in KJ/m².

2-Surface hardness test: this was determined using durometer hardness tester from type shore D, (**hardness tester-th 210, time group Inc. Italy**) which is suitable for acrylic resin material. The instruments consist of blunt-pointed indenter 0.8mm in diameter that tapers to a cylinder 1.6mm. The indenter is attached to a digital scale that is graduated from 0 to 100 units; measurements were taken directly from the digital scale reading. Five measurements were done on different areas of each specimen (the same selected area of each specimen), and an average of five readings was calculated.

3- Surface roughness test: the profilometer device (**Surface roughness tester SRT-6210, England**) was used to study the effect of aluminum silicate filler reinforcement on microgeometry of the test surface. This device is supplied with surface analyzer (sharp stylus) made from diamond. Maximum distance that can be move is 11mm. Two measurements were done on different areas of each specimen (the same selected area of each specimen), and an average of two readings was calculated.

3. Results

FT-IR spectrum obtained from aluminosilicate composite indicated that the peak at 1,05699 confirming the formation of Al-O-Si bonds, and a peak at 443.63 cm⁻¹, which is due to O-Si-O bending vibrations. The wide IR band in the range of 3,000-3,800 cm⁻¹ is due to the stretching vibration of H₂O molecules. Figure 2.

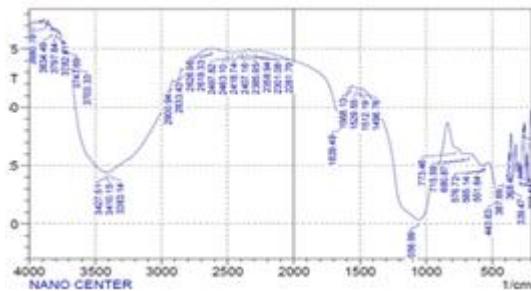


Figure 2: FTIR of aluminum silicate

The absorption bands of modified aluminosilicate fillers has almost all the absorption peaks present in TMSPM, in addition to the peaks present in the aluminosilicate fillers. FTIR spectra of modified aluminosilicate showed adsorption at $1,051.20\text{cm}^{-1}$ (C-H), 1687 cm^{-1} (C=O), 1153cm^{-1} (C-O). This clearly indicated the existence of chemically bonded TMSPM on the aluminosilicate fillers surface as shown in Figure3.

FT-IR silanated Aluminosilicate added to PMMA showed change the shape of adsorption peak at 1627 cm^{-1} (C=C) of PMMA. As shown in Figure 4.

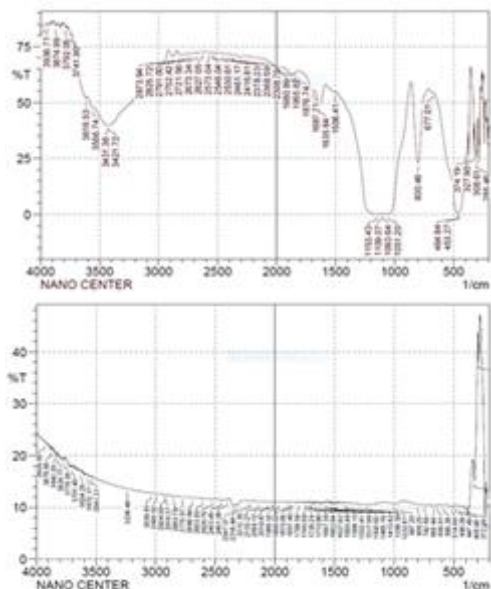


Figure 4: FTIR of silanated aluminium silicate added to polymethylmethacrylate methacrylate

In this study, the comparison was done between unmodified acrylic resin sample (control group) and modified acrylic resin containing 3%, 5% and 7% silanized aluminum silicate composite particles. All data were subjected to a one-way analysis of variance (ANOVA) and followed by multiple comparisons by Bonferroni test method.

Table-2 shows the mean and standard deviation values of impact strength, surface hardness, and surface roughness for all experimental groups. For impact strength test, the highest mean value was (0.288 ± 0.02) for specimens containing 3% Al_2SiO_5 only. While for surface hardness test surface roughness test, there is marked increase in the mean values as the concentration of fillers increase. In ANOVA test, there is a highly significant differences among all tested group in all test ($p\text{-value}=0.000$). In table-3 Multiple

comparisons (Bonferroni test) among all tested group for impact strength mean values. The results shows a highly significant difference ($p\text{-value}=0.00$) between mean values of each two tested group.

In table 4, the results of surface hardness test shows a no significant difference between specimens contain 5% Al_2SiO_5 and specimens contain 7% Al_2SiO_5 only.

For surface roughness test, Table-5 mention that there is a non-significance differences between mean values of a specimens contain 3% Al_2SiO_5 and control group (0% Al_2SiO_5). And between groups contain 5% Al_2SiO_5 and groups contain 7% Al_2SiO_5 .

4. Discussion

The fracture property of poly methyl methacrylate acrylic denture base material is not uncommon so studies to improve the strength of this material by addition of fillers still continued. The Addition of inorganic filler to acrylic denture base in different concentrations to improve its mechanical properties has been studied by many authors^(10, 11)

One of these inorganic fillers alumina and silicate particles.^(21,22) For the aluminum silicate composite material it was proved their biocompatibility and its color white powder not affect esthetic of acrylic denture base.⁽¹⁵⁾

In this study the results of impact strength revealed that a highly-significant difference between control groups and the modified test groups. there is a slight increase in the mean value of 3% compared to control group this attributed to interaction between the silanated aluminum silicate fillers with PMMA matrix. As the matrix acts as impact modifier that it slight increases the ductility of PMMA matrix.

generally the mean values of impact strength decrease with increase the filler content in 5% and 7% test groups compared to control, these results in agreement with Khalaf in 2013⁽²³⁾ & Ronak and Pranav in 2016.⁽²⁴⁾ Decreasing the mean values of impact strength with increase in concentrations of fillers.

The results of hardness test showed that an increase in the mean values with increase the amount of filler and show a highly significant difference between control and 3%, 5% and 7% Aluminum silicate test groups respectively, the possible explanation may be due to the aluminum silicate particles harder than acrylic polymer so increase inorganic filler per unit area of acrylic resin gives more resistance to indenter penetration and more hardness values obtained. Similar results were obtained by different authors who worked on different inorganic micro fillers that added to polymethyl methacrylate denture base material^(25,26)

About the surface roughness the results showed a highly significant increase in surface roughness with increase the filler content this may be due to differences in particle roughness and microstructure of aluminum silicate compared to heat cure acrylic matrix these results agreed with Khalaf, 2013⁽²³⁾ and not agreed with Abdulhamed &

Mohammed in 2010⁽²⁷⁾ and Makarem 2015⁽²⁸⁾ who revealed that the surface roughness of acrylic denture base was not significantly increased with the addition of Al₂O₃. So these differences may be regarded to differences in filler particle type and the method of mixing in both previous studies compared to this study.

5. Conclusion

Within the limitation of this study, it may be concluded that the addition of 3% Aluminum silicate to heat cure acrylic denture base material slightly increase but not significantly improve the impact strength of acrylic denture base. The impact strength decrease with increase particle percentage more than 3% compared with non-reinforced group.

Surface hardness and surface roughness of heat cure acrylic denture base increased with increased the amount of aluminum silicate particles.

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Table 2: Mean, Standard deviation and ANOVA test of all tested sample

Groups		impact strength test		surface hardness		surface roughness	
concentration	N	Mean	Std. Dev	Mean	Std. Dev	Mean	Std. Dev
0% control	10	0.205	0.043	72.382	1.723	2.194	0.209
3% Al ₂ SiO ₅	10	0.288	0.020	75.576	1.395	2.682	0.732
5% Al ₂ SiO ₅	10	0.156	0.012	77.91	2.248	3.336	0.097
7% Al ₂ SiO ₅	10	0.12	0.026	79.594	1.183	3.624	0.247
ANOVA test	F-test	66.449		34.345		25.493	
	sig	0.000		0.000		.000	

Table 3: Multiple comparisons (Bonferroni test) among all tested group for impact strength mean values KJ/M²

Sample	sample	Mean Difference	Std. Error	Sig.
0% control	3% Al ₂ SiO ₅	-0.083*	0.0126	.000
	5% Al ₂ SiO ₅	0.049*	0.0127	0.003
	7% Al ₂ SiO ₅	0.085*	0.0126	0
3%	5% Al ₂ SiO ₅	0.132*	0.0123	0
	7% Al ₂ SiO ₅	0.168*	0.0127	0
5%	4.00 Al ₂ SiO ₅	0.036*	0.0129	.043

*The mean difference is significant at ≤ 0.05 level.

Table 4: Multiple comparisons (Bonferroni test) among all tested groups for indentation hardness mean values

Sample	Sample	Mean Difference	Std. Error	Sig.
0% control	3% Al ₂ SiO ₅	-3.194*	.754	.001
	5% Al ₂ SiO ₅	-5.528*	.754	.001
	7% Al ₂ SiO ₅	-7.212*	.754	.000
3%	5% Al ₂ SiO ₅	-2.334*	.754	.023
	7% Al ₂ SiO ₅	-4.018*	.754	.000
5%	7% Al ₂ SiO ₅	-1.684	.754	.191

*The mean difference is significant at ≤ 0.05 level

Table 5: Multiple comparisons (Bonferroni test) among all tested group for surface roughness mean values μm .

Sample	Sample	Mean Difference	Std. Error	Sig.
0% Control	3% Al ₂ SiO ₅	-0.488	0.181	.063
	5% Al ₂ SiO ₅	-1.142*	0.180	.000
	7% Al ₂ SiO ₅	-1.430*	0.182	.000
3%	5% Al ₂ SiO ₅	0.654*	0.180	.005
	7% Al ₂ SiO ₅	-0.942*	0.183	.000
5%	7% Al ₂ SiO ₅	-.288	0.181	.716

*The mean difference is significant at ≤ 0.05 level