AL-AZHAR
Assiut Dental Journal

The Official Publication of The Faculty of Dental Medicine, Al-Azhar Assiut University, Egypt

AADJ, Vol. 5, No. 1, April (2022) — PP. 1:9

ISSN 2682-2822

Effect of Addition Rubber Modified Montmorillonite on Some Properties of Heat Cured Polymethyl Methacrylate Resin

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Codex : 01/2022/04

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ABSTRACT

Aims: The goal of this research was to create a composite of rubber modified-montmorillonite (MMT) and heat cured Polymethyl Methacrylate (PMMA) resin to improve some of the mechanical and physical properties of heat cured (PMMA). **Subjects and Methods**: A total of 120 specimens were made and divided into four groups, (n=30) for each group. Group A, control group made of conventional heat cured PMMA with no reinforcement. Specimens of the other groups were reinforced by the rubber modified MMT composite with different percentages (2%, 4% and 8%) by weight. Group B (2% composite), group C (4% composite) and group D (8% composite). All specimens were evaluated as regard impact strength, transverse strength, water sorption, hardness, and roughness. **Results:** The results of this study revealed that, impact strength values of acrylic resin denture base were increase after the addition of rubber MMT –composite at any ratio. After addition of 2% rubber MMT-composite the flexure strength was nearly similar to that of control group. While, a significant reduction occurred after adding 4% and 8% composites. Water sorption was significantly increased by the addition of 4% and 8% of rubber –MMT composite ($P < 0.05$). There was a significant decrease in the micro hardness after the addition of 4% and 8 % composite, while insignificant decrease occurred with 2% ratio. As regards to surface roughness there was significant increase in the surface roughness values after addition of rubber –MMT composite for all ratios ($P < 0.05$). **Conclusion**: The most favorable properties were observed by the addition of 2% rubber –MMT acrylic composite to the heat-cured acrylic resin.

KEYWORDS

Rubber, modified-montmorillonite (MMT), acrylic resin, mechanical and physical properties, heat cured Polymethyl Methacrylate (PMMA).

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INTRODUCTION

PMMA resin is the most common material used for making denture bases,[(1)] It has several advantages such as ease of fabrication, repair, relining possibility, affordable cost and good appearance.[(2)]

However, PMMA denture base material has drawbacks such as insufficient surface hardness, low flexure and impact strength. Clinically, failure of complete or partial denture prosthesis is most likely in the shape of fracture either due to fatigue or impact forces with a reported rate of 68% after 3 years of usage.[3]

The low flexure and impact strength are still the most significant disadvantages of PMMA resin which need to improve. Recent advancements in the Flexural strength (bending strength or modulus of rupture) is defined as the force per unit area at the instant of fracture in a test specimen subjected to flexural loading.[4]

Impact strength can be defined as the capacity of a material to withstand a suddenly applied load or stress. These flexural fatigue or impact forces resulting in the concentration of stress around micro-cracks formed within the denture base material because of the repeated submission of in vivo forces. Continuous force leads to crack propagation, which, in turn, reduces the strength of the denture base and finally results in a midline fracture.[5]

Hardness is one of the surface property which is defined as the resistance of a material to plastic deformation.[6] Vickers hardness numbers indicate that acrylic polymers are relatively soft in comparison to alloys. This predisposes the acrylic denture base to wear, caused by abrasive foodstuffs and particularly abrasive dentifrice cleanser.[7]

Roughness is another property related to the surface which is of clinical importance since it may affect plaque accumulation and staining.[8, 9]

Patients dislike rougher denture base surfaces because they cause discoloration, plaque accumulation, and bacterial and fungal adhesion.[10]

Therefore the material should possess a smooth, polished surface so that plaque accumulation is minimized or avoided.[11]

It has been largely reported that the combination of nanofillers to a pure polymer matrix increases some appropriate material properties, like the flexural and Impact strength.[12]

Recently, the combination of filler particles with nanometric dimensions such as clay, silver, zirconium oxide, and calcium carbonate into dental resins has attracted great attention to get a new Polymer nanocomposite that bypass the drawbacks of PMMA. Wear resistance, retention, elastic modulus, flexural strength, tensile strength and reduced polymerization shrinkage have been improved by the addition of this particles.[13] [14] [15]

Organically modified montmorillonite (OMMT) is one of the nanomaterials incorporated into polymer due to its excellent esthetics and biocompatibility. It is used to derive nanocomposites with high hardness, fatigue, tensile and impact strength. However, there are insufficient data evaluating the flexural strength and polymerization shrinkage of the resulting nanocomposite.[4]

Rubber modified-montmorillonite (MMT) was used with certain ratios to reinforce polymethyl methacrylate denture.[16] Unfortunately, there is a shortage in previous studies relating to using different ratios of this composite and its effects on the other physical and mechanical properties.[17]

Therefore this research was designed to study the effect of adding rubber modified-montmorillonite (MMT) with new ratios (2%, 4%, and 8%) by weight on transverse strength, impact strength, water sorption, micro-hardness and surface roughness of heat cure denture base material.

The research hypothesis was that the incorporation of a new ratios of rubber–MMT composite with heat cured PMMA resin would affect some of its mechanical and physical properties.

MATERIAL AND METHODS

A rubber modified-montmorillonite composite, is composed of Montomorillonite (MMT)* (hydrous alumino-silicate clay composed of units made up of central alumina sheet sandwiched between two

* Dentstply International Inc., York division, U.S.A.

silica sheets).[18] and Amine- terminated butadiene acrylonitrile rubber materials.[19] The ratio was 1:1

The methods of formation the rubber modified-montmorillonite (MMT) was previously reported[16].

A pre-weighted amount of rubber-MMT composite was added to the powder of conventional heat cured PMMA** at different ratios, 2 %, 4 % and 8 % by weight.

The specimens were processed according to the specification of each test. (Impact strength, flexural strength, and water sorption) and were divided into four equal groups, 30 specimens in each group as follow.

Group A: control group only conventional heat cured PMMA resin.

Group B: PMMA resin reinforced with 2% rubber-MMT

Group C: PMMA resin reinforced with 4% rubber-MMT

Group D: PMMA resin reinforced with 8% rubber-MMT

The test specimens were made by inclusion of the conventional heat-cured PMMA to the selected ratios of the rubber-MMT material. The proper monomer to polymer ratio was 1: 2.5 by weight and 1% of benzoyl peroxide (initiator) was added to the powder.

The specimens were cured in the same curing cycle. The specimens were smoothed and polished by using a silicon carbide papers grades 240-600 and pumice and slurry. It was stored in distilled water at 37 ± 1°C for 48 hours before testing.

Methods

Impact strength test:

The impact strength test specimens were done according to British Institute specification no 771. (British Standard Institute Specification No.771; 1984).

Each specimen had the dimension of 75 mm (length) x 10 mm (width) x 10 mm (thickness) with standard notch of 2mm (depth) at mid-span.

The impact strength was determined by the use of Izod Impact Tester (Brooks, Model IT 14 Press 1,68 Kg) The energy (E) required for breaking the test specimens were calculated from the following formula [20].

$$E= W R (\cos \beta - \cos \alpha) - L$$

E: Energy required for breaking the test specimen in joule.

W: Weight of pendulum in Newton.

R: Distance from the axis of rotation to the center of gravity of the pendulum, in meter.

β: Angle of the rise of the pendulum after breaking the specimen.

α : Angle of fall of pendulum L: Loss of energy due to friction, in joule.

Where E was dividing on the cross-section area at the notch.

Transverse strength test:

The transverse test specimens were made in the shape of a flat strip in accordance with A.D.A. Specification No.12. [20], with a dimension of 65mm (length) x 10 mm (width) x 2.5mm (thickness).

The number, preparation, curing, finishing and storage of specimens were done as described for impact strength test.

The test was achieved by universal testing machine (Instron 3600 series, USA universal testing machine) **Fig. (1)** The transverse strength was calculated from the following formula [21]

** Acrostone Dental factory, England.

$$S = \frac{3\,P\,I}{2\,b\,d}$$

S: Transverse strength, in MPa.

P: Maximum load before fracture, in Newton.

I : Distance between supports, in mm.

b : Width of the specimen, in mm. d : Thickness of the specimen, in mm.

Water sorption test:

Specimens were made in the shape of disk in accordance with A.D.A. specification No. 12. [22], 50 mm. in diameter and 0.5 mm in thickness. The number, preparation, curing, finishing and storage of specimens were done as described for impact strength test.

The resin disks were dried in a desecrator at 37 ± 2°C for 24 hours, removed to a similar desecrator at room temperature for one hour then weighted by using a high accuracy balance instrument (Sartorius MCI Research RC 210D,Sartorius AG, Gottingen, Germany) until the constant to an accuracy of 0.2mg.

This cycle was repeated until the reduction in weight of each disk is not more than 0.5 mg. in 24 hours period. The disks were then immersed in distilled water at 37 ± 1°C for 7 days, then the disks were removed from the water and wiped until it is free from visible moisture and weight. The role of water sorption was calculated as follow for each disk.

$$\text{Water sorption (mg/cm2)} = \frac{(m1) - (m)}{(a)}$$

(m1) : Mass after immersion.

(m): Conditioned mass.

(a): Surface area

Micro-hardness test:

Vickers hardness tester (ZwicRoell, west Midlands, England) was used to determine the specimens' hardness. Testing was done under an applied load of 10 gm. for 20 seconds. Using an average of five readings for each strip.

Surface roughness test:

Surface roughness was measured in accordance to Zissis et al.,[11] [23] The method used is to scan a diamond stylus ((Posi-tecter, SPG, Deflesko Co., USA)) across the surface under constant load and computed the numeric values representing the roughness of the profile as (Ra). The (Ra) parameter describes the overall roughness and can be defined as the arithmetical average value of all absolute distances of the roughness profile from the center line within the measuring length. For each specimen five reading were carried out, and the mean of these readings was used for the statistical analysis.

For each test, range, mean and standard deviation (SD) values were calculated and analyzed using One way ANOVA followed by pair-wise Tukey's comparison tests on Past softwar (Natural history Museum) version 2.17 to determine the significance detected between subgroups.

Fig. (1) Universal Testing Machine

RESULTS

Table (1) Shows the mean and SD of the tested groups in comparison with the control group for all five tests.

Table (1) *Mean (SD) values of different studied groups as regards Impact strength, transverse strength, water sorption, Vickers hardness and roughness. *Significant, P < 0.05*

	Group- A (control)	Group- B (2%)	Group- C (4%)	Group- D (8%)
Impact strength	3.194* (0.487)	4.046* (0.532)	5.056* (0.468)	5.927* (0.132)
Transverse strength	90.442 (1.496)	90.388 (1.056)	89.674 (0.812)	89.42 (0.608)
Water sorption	1.15 (0.045)	1.22 (0.0452)	2.037 * (0.343)	2.437 * (0.055)
Hardness	42 2.828	40.8 2.828	37.2 * 1.166	33.4 * 2.059
Roughness	37 (2.28)	53 * (3.162)	64.8 * (3.544)	78.4 * (4.317)

Impact strength:

It was evident that there was significant increase in the impact strength with increasing the rubber (MMT) ratios, $P < 0.05$.The highest mean value was observed in group D ($5.927 J/mm^2$) while the lowest mean value was in the control group ($3.194 J/mm^2$).

Transverse strength:

The highest value recorded for transverse strength was recorded for the control group (90.442 MPa). While the lowest mean value was recorded for group D (89.42 MPa). The transverse strength was decreased as the ratio of rubber (MMT) increased but without any significant difference Fig. 2

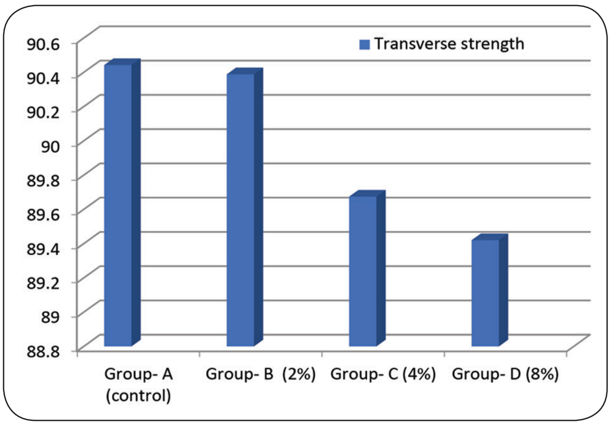

Fig. (2) Comparison between the Transverse strength of tested groups.

Water sorption:

It was noted that the highest mean values of water sorption (%) was recorded for group D (2.437%), while the lowest value recorded for control group (1.152%). There was significant differences between the mean values of water sorption of groups (C and D) and the control group. And between C and D as compared to group B, while the difference between group B and the control group was insignificant

Micro-hardness:

On the other hand the highest mean value of hardness was recorded in the control groups (42 VHN) while the lowest mean value was observed in group D (33.4 VHN). It was evident there was a significant differences between groups (C & D) and the control group. And between groups (C & D) as compared to group B, while the difference between group B and the control group was insignificant. Fig 3

Surface roughness:

Also the results showed that the highest surface roughness value was observed after addition of 8% rubber- MMT composite (78.4 µm). But the lowest mean value was recorded in the control group (37µm). There were significant differences in the surface roughness values after addition of composite at any ratio in comparison with the control group, P< 0.05, Fig 3.

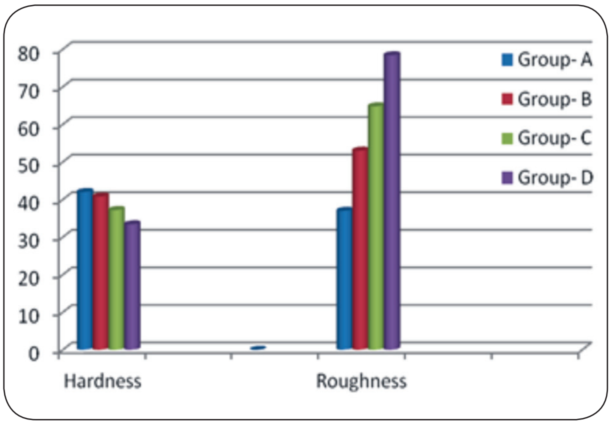

Fig. (3) Comparison between the hardness and roughness of tested groups.

DISCUSSION

PMMA is the most commonly material used in the manufacturing of removable dental prosthesis. Despite its popularity, it is still far from ideal in fulfilling the physical and mechanical requirements of dental prosthesis.(4)

Many studies have been conducted in order to improve the physical and mechanical properties of acrylic denture bases. In this research we demonstrated the effect of adding new ratios of rubber modified-montmorillonite composite on some tested properties of acrylic denture base material including, impact strength, transverse strength, water sorption, hardness and roughness. So the results of this study support our hypothesis .

The results showed that incorporation of rubber-MMT composite to the heat cured PMMA increased the impact strength. This may be due to the fact that rubber absorbs a greater amount of energy at high strain rate before it fractures. This result is in line with previous studies that reported a high impact strength was developed by using a butadiene-styrene rubber.(24,25)

Although the results of the present study showed that there was reduction in the transverse strength after increasing the rubber –MMT percentage, it was noticed that no significant reduction occurred after addition of 2% rubber. However according to ISO 1565, these results are considered an acceptable strength values as they were all more than 65mpa.(4)

The reduction in the transverse strength may have been due to the plasticizing effect of rubber –MMT that affect the strength properties of the acrylic composite. (26,27) This finding in agreement with previous studies that reported that the flexure strength decreased as the rubber content in the PMMA denture base increased.(28,29) On the other hand these results disagreed with other studies.(30,31)

Dimensional stability of dentures during processing and function is of considerable importance as regards to fit and satisfaction to the patient. (4) This study revealed that the water sorption increased after the addition of rubber- MMT composite however this increase was only significant when the percentage increased more than 2% .

This finding is in agreement with other studies,(26,16) and it may be attributed to the swelling and hydrophilic characteristics of MMT. These results disagreed with the study that reported that the water sorption value was decreased by adding of the rubber to PMMA.(32)

The result of the present study revealed that there was an inconsiderable reduction in micro-hardness after addition of 2% rubber but a significant decreasing occurred when the composite ratios were increased more than 2%. This finding may be due to increasing the water sorption resulted from addition of high ratio of rubber content.[26] On the other hand, this finding disagrees with the results of other research.[33]

In our research, the most rough surface was recorded after incorporation of 8 % rubber (MMT) while the least rough group was the control group . The roughness increased by increasing the rubber ratios. These results were coinciding with previous studies.[11, 17]

Future research should be done to demonstrate the effect of addition of rubber modified-montmorillonite composite to heat cure PMMA with different ratio on other properties.

CONCLUSION

Within the limitations of the current study, we can conclude that:

1. Impact strength of heat cure PMMA resin increased after reinforcement with the rubber modified-montmorillonite composite in different ratios.
2. After addition of 2% composite, the transverse strength was nearly similar to that of control group. While insignificant decrease occurred with 4% and 8% of composite ratios.
3. The water sorption values were significantly increased only by addition of 4% and 8% percent of rubber –MMT composite.
4. Increasing the rubber MMT composite ratio within PMMA decreased the hardness of acrylic resin denture base and increased its surface roughness
5. The most favorable properties were observed by the addition of 2% rubber –MMT acrylic composite to the heat-cured acrylic resin.

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AADJ, Vol. 5, No. 1, April (2022) — PP. 9

تأثير اضافة طفلة المنتمريلونايت المعدلة بالمطاط علي بعض الخواص لمادة رانتج الأكريل حرارية الطبخ

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الملخص:

الهدف: تأثير اضافة طفلة المنتمريلونايت المعدلة بالمطاط بتركيزات مختلفة (2%، 4% و 8%) بالوزن من مادة طفلة المنتمريلونايت المعدلة بالمطاط علي الخواص الأتية لمادة رانتج الأكريل حرارية الطبخ (خاصية قوة الصدم-خاصية قوة الانحناء المتعرض-خاصية امتصاص الماء-خاصية الصلابة-خاصية الخشونة.

الطرق والاساليب: تم تحضير 90 عينة طبقا للموصفات العالمية الخاصة بكل تجربة الي اربع مجموعات لدراسة الخواص السابقة. مجموعة (أ) مجموعة تحكم مصنوعة من الأكريل الحراري، مجموعة (ب) تركيز 2% ، مجموعة (ج) تركيز 4%و مجموعة (د) تركيز 8%. كل مجموعة تحتوي علي 30 عينة لقياس الخمس الأختبارات المذكورة باستخدام أجهزة القياس الخاصة بكل تجربة. وقد تم تحليل جميع البيانات إحصائيا من خلال طريقة واحدة ANOVA ، توكي آخر اختبار مخصص وعينة اختبار الاقتران.

النتائج: زيادة قوة الصدم بزيادة النسبة المضافة من اضافة طفلة المنتمريلونايت المعدلة بالمطاط و نقص قوة الانحناء المستعرض والصلابة بزيادة نسبة طفلة المنتمريلونايت المعدلة بالمطاط وزيادة خاصية امتصاص الماء وخشونة السطح بزيادة النسبة المضافة من اضافة طفلة المنتمريلونايت المعدلة بالمطاط

الخلاصة: ان أفضل الخواص كانت للاكريل بعد اضافة نسبة 2% من طفلة المنتمريلونايت المعدلة بالمطاط

الكلمات المفتاحية: المطاط، المنتمريلونايت المعدلة،الاكريل، رانتج الأكريل حرارية الطبخ، الخواص الكميائيه والفزيائيه.