Impact strength behaviour of PMMA denture base through addition of different nanoparticles after immersion in some nutrition liquids

Zaynab N. Rasheed and Samah M. Hussain
Applied Science Department, Technology University, Baghdad, Iraq
E-mail: znraziky@yahoo.com

Abstract
Poly methyl methacrylate PMMA polymer could be considered the main material that used mostly in the recent years in denture base fabrication. It commonly known by it is poor strength properties such as low impact strength. The aim of the present research was to enhance the performance of PMMA denture base through the addition of two kind of nanoparticles (nano particles that selected from artificial and natural sources). Nano-particles from both Al2O3 and crushed peanut Peel were used for comparing purposes. Various weight fraction used in this study for both kinds of the additive (1%, 2% and 3%). Moreover, in this work a study and evaluation in impact strength (I.S.) value were done before and after immersion. The new prepared nanocomposite in three different liquids (mineral water, natural lemon juice and Pepsi) immersed during three specific time (10, 20 and 30 min), all tests completed at room temperature. It was found that the impact strength value before immersion decreased gradually during reinforcement with both type of nanoparticles except when using 3% of Peanuts Peel nanoparticles. Also, it was found after immersion pure PMMA in the three different liquid that the value of I.S. decreased. When immersion the prepared sample inside mineral water, it was noted that using Al2O3 as reinforcement the determined value decrease with increasing the weight fraction different from the Peanuts Peel. The obtained results showed that immersion these samples in naturel lemon juice increased the value of impact strength gradually with the time. I.S. value decreased while immersion nanocomposite of Al2O3 with Pepsi, while an obvious increase was clear with nanocomposite of Peanuts Peel with the immersion time.

Key words
PMMA, impact strength, Al2O3 nanoparticles reinforcement, peanuts peel nanoparticles reinforcement, immersion in nutrition liquids.

Article info.
Received: Jun. 2018
Accepted: Dec. 2018
Published: Jun. 2019
Introduction

In denture application removable teeth are basically used to take parts instead of missing teeth [1]. So, synthetic denture base mostly prepared by heat curing technique using Poly methyl methacrylate (PMMA) since 1940 the primary family of acrylic resin [2]. PMMA approved to be the universal versatile polymer in denture base [3]. The reason behind this selection that PMMA material has simple fabrication process also has some properties like low cost, light weight, colour matching ability, excellent biocompatible material, stability in oral environment, easy finishing and polishing technique [4, 5]. However, PMMA possess insufficient value in mechanical strength when used alone. Moreover, during sudden accident or when a high mastication force applied by a patient on the denture base the result is the easy failure of the prepared base [6]. Yet, these disadvantages could be overcome through the addition of some reinforcement, these problems includes low in strength and brittle [4, 7, 8]. Generally, fractures in denture base happen due to heavy occlusal forces and prolonged use. Denture fracture usually accompanied by fatigue and impact failure, while for mandibular dentures, impact responsible of 80% of fractures and involves extensive repair costs by countries [9]. Only UK had to repair more than 1 million denture base due to poor properties of both impact and fracture strength in 1997. More understanding and definition of fracture mechanism are required, also the proper procedure that to enhance these faults in the material utilise are highly essential in dental world [10]. In addition to all the above, some fracture occurs may be related to design imperfection, material fabrication and choice [11].

Essentially, impact strength and fracture toughness could be considered as the most properties required with high performance in order to consider the used denture base resin are in excellent condition for long term use [12]. Ahmed and Ebrahim declared that almost 70% of used dentures were broken and damaged during the first three years, this study used a compression of ten types of denture base resins [13]. Fracture normally happen in many cases like when the user applies high mastication force between Upper jaw and mandible jaw [14]. Moreover, deformation effect could occur during the time due to biting and mastication force [15-17].

Basically, in the recent decade many researchers presented studies to improve the general properties of denture base materials through the reinforcement of different fillers in to PMMA [18, 19]. These various additive includes fibres [20, 21], nano particles [22-25] and whiskers[26], etc.
Nanoparticles addition may improve the mechanical properties of PMMA because of the high surface area-to-volume ratio. So, this ratio enhance the performance of nanoparticle through the better interfacial interaction of with the PMMA resin [27]. The improvement of the nanocomposite mechanical properties critically depends on the type of incorporated nanoparticles used for the reinforcement, specially the size the type and even the distribution. The concentration and the interaction of these nano-reinforcement with the resin matrix are also important for better properties [28]. Nano particles integrate with the polymeric matrix to improve most of the mechanical properties such as the rigidity, fracture toughness and other functional properties of the new nanocomposite [29]. Different researcher used Al2O3, Zr2O3, and SiO2 as nanofiller in their studies [13, 27]. In dental composite and interfacial silane, reformulation nanoparticles were greatly used [27].

However, there is still no research presents an experiment results on nanoparticles reinforcement effect on the impact strength of PMMA resin before and after immersion in food liquids. Hence, it was the main aim in this research to study the ability of some artificial and natural nanoparticles to improve the value of impact strength of PMMA resin. These additive were investigated to proof whether it could be considered as promising for reinforcement PMMA polymer resin or not.

**Experimental part**

**Technique of samples preparation**

**A-Materials**

In the current study PMMA (methyl methacrylate) used as the only resin and reinforced through different kind of nanoparticles to prepare our nano-composites:

1. (Al2O3) Nanoparticles.
2. Peanuts Peel Nanoparticles.

**B-Mould and sample preparation**

The mold used to prepare test sample was made of glass with fix dimensions (15 cm × 10 cm × 0.4 cm) and covered with a glass plate to provide smooth sample surface.

The Vertex™ Castavaria was used to prepare the specimens of the PMMA composite materials. The standard proportion for mixing weight ratio is usually for cold cure acrylic resin (50% to 50%) from polymer powder and monomer liquids (MMA). PMMA is moldable for a long period of time, where the liquid of (MMA) was poured in clean and dry container (glass beaker), followed by a slow addition of dry powder of polymer to form the final resin. Then, the prepared mixture was stirred at room temperature continuously using hand lay-out technique until the dough stage. Then it was poured in the center of the glass mould with maximum time about (3 min). The internal wall must covered with thin layer of Vaseline to avoid sticking between cast material and Mould wall, also to avoid the formation of bubbles inside the mixture a slow and continues mixing was applied. This mixture was left at room temperature continuously using hand lay-out technique until the dough stage. Then it was poured in the center of the glass mould with maximum time about (3 min). The internal wall must covered with thin layer of Vaseline to avoid sticking between cast material and Mould wall, also to avoid the formation of bubbles inside the mixture a slow and continues mixing was applied. This mixture was left at room temperature for (1 hour) for solidification. The casting sheet was released from the mould and placed in an oven at (55 °C) for another (1 hour) to post cure the considered sample sheet.

The Prepared nano-composite specimens were made from PMMA polymer reinforced with nanoparticles of both (Al2O3 and Peanuts Peel). The reinforcement percentage was made by weight friction between (1 %, 2 % and 3 %) using hand lay- up technique with same size glass mold that used before. To be noted that the particle size of Al2O3 was between (30-35) nm from
Changsha Santech Co. according to the manufacturer. While the peanuts peel nano-particles were made in some simple steps: first clean the peel with normal water and dried at room temperature then manual hammering performed to minimize the particle size. Lastly the obtained crushed peanuts peel put inside a mechanical nano-grinding ball for about 2 hours to decrease the size until nanometer scale. Also, to ensure the particle precise size after grinding particle size analyzer (90-plus) used in this work. Fig.1 present the average particle size finally obtained for the utilize samples, the obtained effective diameter for the utilized nanoparticles was around (1.576) µm.

![Fig.1: The lognormal size distribution.](image)

The prepared (PMMA) and reinforced nanoparticle (Al₂O₃, Peanuts Peel) must be mixed at room temperature continuously by using hand lay-out mixing to obtain the homogenous mixture. All mixtures consist of PMMA powder and the added nanoparticles must mix with MMA liquid resin until reach the dough stage. Table 1 explains the 6 different mixture prepared for this research with full details.

It shown in Fig. 2 the two type of the prepared Nanocomposite (PMMA + Al₂O₃) and (PMMA + Peanuts Peel) with the three weight fraction (1%, 2%, 3%). It is clear from the figure that the increase of weight fraction of Al₂O₃ changes the color from transparent pink to light pink, while adding more from the Peanuts Peel nanoparticles change the sample color to beige. Finally, the prepared nano composites plates were cut into the mentioned dimension above based on ASTM standard for the impact test. After preparing impact samples immersing procedure were applied in three nutrition liquids (mineral water, natural lemon juice, Pepsi) for different period of time (10, 20 and 30 min).

| Matrix       | Sample code | Addition nanoparticles          |
|--------------|-------------|---------------------------------|
| PMMA powder  | S1          | (No additive)                   |
| liquid + MMA | S2          | 1%wt Al₂O₃                      |
| PMMA powder  | S3          | 2%wt Al₂O₃                      |
| MMA powder   | S4          | 3%wt Al₂O₃                      |
| Peanuts Peel | S5          | 1%wt peanuts peel               |
|              | S6          | 2%wt peanuts peel               |
|              | S7          | 3%wt peanuts peel               |

Table 1: Symbols of each type of material under investigation.
Impact test

Charpy impact test (I.S.) was carried out in this work in order to evaluate the value of fracture toughness before and after the reinforcement process on PMMA resin. Also, more investigation on the behavior of this test was taken through immersion the prepared nanocomposite samples in three specific liquids for fixed time. The tested samples cut according to ISO-179 standard, the dimension of the tested samples were (55 mm *10 mm *4 mm) and kept at room temperature (25 °C). The basic principle of Charpy impact test is to determine the amount of energy absorbed by a material sample during the fracture, which refer to material toughness. Fig. 3a shows Charpy impact instrument (Testing Machines INC. AMITYVILLE, New York) used, where a Pendulum of energy (5 Joul) used on the utilized samples. While Fig. 3b present the prepared nano-composite samples in different situation after cut. The impact strength is calculated from the following relation [30]:

\[
\text{Impact Strength (I.S)} = \frac{\text{Energy of fracture (KJouI)}}{\text{Cross – sectional area (m}^2\text{)}}
\]
Results and discussion

Impact strength had a huge interest in the dental application especially in the manufacturing of the synthetic denture base. As mentioned before that most of denture failure happened due to impact strength failure [9]. The basic principle that the impact test work with is the absorption of the kinetic energy from the swinging hammer as shown previously in Fig. 3. The tested sample is supported at its side in a way that the fracture must take place in the middle of the piece. Some of the kinetic energy absorbed from the spacers while the other is responsible of the sample fracture. The fracture energy is the value use to determine the impact strength I.S. Generally, the failure in the sample occurs due to applied stress, under a dynamic quick stress the material tend to behave as brittle rather than ductile [31, 32]. Therefore, the reinforcement effect on the prepared composite is to increase the energy required to break the sample under investigation [33]. This energy represented by the value of impact strength (I.S.). Impact strength before immersion process measured for (pure PMMA, Nanocomposite reinforced with first Al$_2$O$_3$ nanoparticles and second reinforced with Peanuts Peel nanoparticles) respectively. Fig.4 indicates the results obtained for all mentioned samples, clearly the reinforcement using the Peanuts Peel nanoparticles gave better result than the other. This could be explain due to the brittle nature that the Al$_2$O$_3$ nanoparticles consist causing the slight decrease in the I. S. value (decreased from the absorbed energy after reinforcement).

![Graph showing the influence of reinforcement on impact strength](image)

Fig.4: Influence of reinforcement of Pure PMMA with (1%, 2%, and 3%) of Al$_2$O$_3$ and Peanuts Peel nanoparticles respectively.

After immersion as indicated in Table 2, the value of impact strength of the pure PMMA was gradually changing with the immersion time specially when using liquids like (mineral water, Pepsi). This could be as a resulting from the liquids effect on the pure PMMA. The liquid work on dissolution of the polymeric material was attributed to failure. Spread the liquid through material components lead to break the bonding and emergence of bubbles that deform the sample easily. While there is slight increase in the impact strength value after immersion with lemon, as the time of immerse increase [34].


Table 2: Impact strength (I.S.) of pure PMMA for (10, 20, 30 min) immersed in (Mineral water, Lemon and Pepsi).

| Immersion liquids | Immersion time (min) |
|-------------------|----------------------|
|                   | 0        | 10    | 20    | 30    |
| Water battle      | 6.7      | 6.1   | 6.4   | 5     |
| Lemon             | 6.7      | 5.8   | 6.5   | 6.5   |
| Pepsi             | 6.7      | 6.7   | 6.4   | 6.1   |

Figs. 5 and 6 illustrate the determined value of the impact strength through immersion in mineral water for the prepared nanocomposite in different type of reinforcement, the fraction weight were (1%, 2%, 3%) for the specific immersion time (10, 20, 30 min). Fig. 5 clearly shows decrease in the impact strength when using Al$_2$O$_3$ as reinforcement material, this decrease related with the increase of the reinforcement weight percentage. This could explain due to the fact that Al$_2$O$_3$ is increasing the brittleness of the nanocomposite due to its nature as ceramic material. Also, the inability of the reinforcement to block the crack propagation resulting in reduction of the absorbed energy required to complete the fracture.

On the other hand, slight increase noted when using the nanoparticles from Peanuts Peel with the samples as illustrated with Fig. 6. This increase related with the increase of the weight percentage through the increase of immersion time. While mineral water inter the material component it decrease the matrix and additive bonding and this lead to increase porosity thus increase absorption of the mineral water and finally increase material plasticity.

![Fig.5: Influence of immersion nanocomposite PMMA + (1%, 2%, 3% of Al$_2$O$_3$) in mineral water for (10, 20, 30 min), respectively.](image-url)
Fig. 6: Influence of immersion nanocomposite PMMA + (1%, 2%, 3% of Peanuts Peel) in mineral water for (10, 20, 30 min), respectively.

Fig. 7 presents the effect of immersion in Lemon for (10, 20, 30 min) with the prepared nano composites using Al₂O₃ nanoparticles. In which the impact strength measured changed differently during the increase of the specific exposure time. This behavior could be explained due to the effect of these liquids inside the nanocomposite samples and the brittle nature of the used additive, which change the material nature more ductile mean thus more absorbed energy will be detect before sample fracture [35]. Interestingly, to be noted that a clear improvement in I.S. value as the weight percentage of the reinforcement increase for the case of using peanuts peel as additive as presented in Fig.8. The increase of the nano-reinforcement help the prepared nanocomposite component to have better stacking during the immersion time, which means increase of the material toughness. These nanoparticles settle inside the polymer material (molecule chains) and work as obstacle to stretch fractions and thus increase the ability to absorb energy [36]. Results obtained using peanuts peel nanoparticles in the prepared samples can be seen in Fig.8. Best value determined for the impact strength was (8.2 kJ/m²) when reinforced with 3% Peanuts Peel and after 30 min of immersion.

Fig. 7: Influence of immersion nanocomposite PMMA + (1%, 2%, 3% of Al₂O₃) in natural Lemon for (10, 20, 30 min), respectively.
Table 3 indicates the impact strength as a function of time with different addition of Al₂O₃. All the Al₂O₃ nanocomposite samples shows a slight decrease in the impact value determined except (1%) which showed a noticeable increase after (10 min) of immersion, this could be as a result of some fabrication default in (1% Al₂O₃) sample that permit the liquid to inter inside the composite sample and increase the plasticity thus increase the absorbed energy which leads finally to higher I.S. before facture. Generally, as the increase in the additive percentage of Al₂O₃ nanoparticles the impact strength determined decreased. The impact strength depend on percentage weight between the matrix and the additive and the linking degree between both. So, for this case the resulted stresses concentrated around the particles location which helps to spreads the cracks and the defects within the composite materials [37]. After that it is clear that with the increase of immersion time the value of impact strength drops and the results manner was not systematic. Table 3 also showed different behavior using the Peanuts peel nanoparticles in the reinforcement work, gradual increase of the impact value were detected. The increase of immersion time leads to increase the I.S. due to the increase of the material plasticity.

**Table 3: Value of impact strength I.S. (kJ/m²) for both nanocomposite PMMA + (1%, 2%, 3%) of Al₂O₃ and peanuts peel respectively during immersion in Pepsi.**

| Samples                  | Immersion time (min) |
|--------------------------|----------------------|
|                          | 0        | 10       | 20      | 30      |
| PMMA                    | 6.7      | 6.7      | 6.4     | 6.1     |
| PMMA+1%Al₂O₃            | 4.6      | 10.6     | 7.2     | 7.3     |
| PMMA+2%Al₂O₃            | 5.6      | 4.5      | 2.3     | 2.3     |
| PMMA+3%Al₂O₃            | 4.5      | 3.5      | 2.4     | 2.8     |
| PMMA+1%P.peel           | 5.7      | 5.8      | 6.6     | 6.7     |
| PMMA+2%P.peel           | 5.5      | 6.5      | 5.8     | 6.8     |
| PMMA+3%P.peel           | 6.8      | 7.2      | 7.5     | 8.0     |

Fig. 8: Influence of immersion nanocomposite PMMA + (1%, 2%, 3% of Peanuts Peel) in natural Lemon for (10, 20, 30 min), respectively.
Conclusion

The effect of reinforcement of pure PMMA polymer with two type of nanoparticle (Al₂O₃ and Peanuts Peel) before and after immersion in three different liquids for specific time were investigated. In general the reinforcement using the natural nanoparticles Peanuts peel showed a better result of Impact strength before and after immersion in comparison with the reinforcement using Al₂O₃. That is mean, these natural additive could replace Al₂O₃ in this field although it consider cheaper and eco-friendly materials. Also, the following conclusion could be drawn.

1. There are noticeable change in pure PMMA after reinforcement, from transparent pink to light pink in the case of Al₂O₃ while to light beige in Peanuts peel case.
2. The impact strength value of PMMA resin before immersion showed noticeable drop after the reinforcement except the case of 3% of Peanuts peel nanoparticles reinforcement.
3. The impact strength value of all the pure PMMA resin decreased after the immersion in all type of liquids and continues as the period of the immersion increases.
4. It was noticed that the prepared nanocomposite reinforced with both type of nanoparticles has slight increased while immersion in Lemon and continue during the increase of immersion time.
5. It was noticed that the effect of immersion in pepsi on the prepared samples with Al₂O₃ mostly decreased from the I.S value except the case of 1% after 10 min which show a higher value (10.5), while using the Peanuts peel nano particles showed a gradual increase.
6. There are no effects observed on the shapes, dimension or color of the samples after the immersion into the different liquids use.

References

[1] J. Jancar, K. Hynstova, V. Pavelka, Comp Sci Technol, 69, (3-4) (2009) 457-462
[2] CPPS. Fernanda, P. Heitor, A. Vieira, LFR. Garcia, Mater. Res., 12, 4 (2009) 415-418.
[3] R. Vivek and R.Soni, International Journal of Dentistry and Oral Health, 1, 4 (2015) 1-3.
[4] V. Rakhshan, Saudi J Dent Res, 6, 1 (2015) 33-44.
[5] X. Xu, L. He, B. Zhu, J. Li, Polym. Chem., 8 (2017) 807-823.
[6] W. Yu, X. Wang, Q. Tang, M. Gue, J. Zhao, J. Med. Behav. Biomed. Mater., 32 (2014) 192-197.
[7] NS. Ayad, H. Elkawash, J Adv. Dent Res, 2, 1 (2011) 33-36.
[8] Z. Han, B. Zhu, R. Chen, Z. Huang, C. Zhu, X. Zhang. Mater. Des., 65, (2015) 1245-1252.
[9] PA. Hari, Kalavathy, HS. Mohammed, Ann Essences Dent., 3, 4 (2011) 7-12.
[10] JF. McCabe, AWG. Walls. Applied dental materials. 9th ed. UK: Blackwell Publishing; 2008.
[11] V. Asopa, S. Suresh, M. Khandelwal, V. Sharm, SS. Asopa, LS. Kaira, Saudi J. Dent. Res. 6, 2 (2015) 146-151.
[12] G.Puri, DW. Berzins, VB. Dhuru, PA. Raj, SK. Rambhia, G. Dhir, AR. Dentino, J. Prosthetic Dent., 100, 4 (2008) 302-308.
[13] M.A. Ahmed, MI. Ebrahim. World J. Nano Sci. Eng., 4, 2 (2014) 50-57.
[14] NV. Asar, H. Albayrak, T. Korkmaz, I. Turkyilmaz. J. Adv. Prosthodont, 5, 4 (2013) 241-247.
[15] AG. Andreopoulos, GC. Papanicolaou. J. Mater. Sci., 22, 9 (1987) 3417-3420.
[16] A.O. Alhareb, Z.A. Ahmed, J. Reinf. Plast. Compos., 30 (2011) 86-93.
[17] D. Gokeliler, S.Erkut, J. Zemek, H. Biederman, M. Mutlu, Dent. Mater., 23 (2007) 335-342.
[18] P.K. Vallittu, Journal of Prosthod, 4 (1995) 183-187.
[19] V. Morino-Maldonado, L.S. Acosta-Torres., F.H. Barcelo-Santana., R.D. Vanegas-Lancon, M.E.Plata-Rodriguez, V.M. Castano, Journal of Applied Polymer Science, 126 (2012) 289-296.
[20] R.J Kane, W.Yue, J.J. Masan, R.K. Roeder, J. Mech. Behav. Biomed. Mater., 3, 7 (2010) 504-511.
[21] V.M. Miettinen, P.K. Vallittu, Biomaterials, 18 (1997) 181-185.
[22] A. Akinci, S.Sen, U.Sen, Compos Part B: Eng., 56 (2014) 42-47.
[23] S.O. Alsharif, H.Bin Md Akil, El-Aziz, Abbas Abd, Z. N Arifin Bin Ahmed, Mater. Des., 54 (2014) 430-435.
[24] Y.Hu, G. Gu, S. Zhou, L. Wu, Polymer, 52 (2011) 122-129.
[25] M.L. Saladino, T.E. Motaung, A.S. Luyt, A. Spinelle, G. Nasillo, E. Gaponetti, Polym. Degrad. Stab., 97 (2012) 452-459.
[26] L. Niu, M. Fang, K. Jiao, L. Tang, Y. Xiao, L. Shen, J. Chen., Tetrapod-loke zinc oxide whisker enhancement of resin composite. J. Dent. Res., 89 (2010) 746-750.
[27] S. Kongo, S. Kalia, A. Celli, J. Njuguna, Y. Habibi, R. Kumar, Prog. Polym. Sci., 38 (2013) 1232-1261.
[28] K.J. Moreno, J.S. García-Miranda, C. Hernandez-Navarro, F. Ruiz-Guillen, L.D. Aguilerama-Camacho, R. Lesso, A. Arizmendi-Morquecho, J. Compos. Mater., 49, 11 (2015) 1345-1353.
[29] C. Qain, X.-Y. Zhang, B.-S. Zhu, K.-L. Lin, J. Chang, X.-J. Zhang, Adv. Compo. Lett., 20 (2011) 13-20.
[30] K. K. Chawla, Composite Materials (Science and Engineering), Springer–Verlag New York Inc., (1987).
[31] Ram, Arie, "Fundamentals of Polymer Engineering", Plenum Press, 1997.
[32] S. Brent.2000. Plastic Materials and Processing. 2nd, Brigham Young University.
[33] J.S Wu, K. Friedrich, M. Grosso, J. Composites, 20, 3 (1989) 223-233.
[34] H.J. Abd Al-Hussien, Iraqi Journal of Science, 56, (3a) (2015) 1963-1952.
[35] V. Curtue., Journal of Applied Polymer Science, 8 (2009) 201-211.
[36] Ashby, Michael, shercliff, Hugh and Cebon, David, (2007), 1st edition, University of Cambridge, Butterworth-Heinemann, Elsevier publications, United Kingdom.
[37] Q.A.H. Aljbouri. (2008), Master Thesis Material Engineering.