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Synthesis and Characterization of PMMA/HAP/MgO Nanocomposite as an Antibacterial Activity for Dental Applications

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ABSTRACT

The current work focuses on the preparation of magnesium oxide nanoparticles using aqueous magnesium chloride salts and sodium hydroxide powder by a simple chemical precipitation method and at an annealing temperature of 700 °C for 6 hours. The structural and morphological properties of the magnesium oxide nanoparticles were investigated using X-ray diffraction (XRD). Polycrystalline cube and using the Scherer equation showed a crystalline size of 17.23 nm. EDS analyzes showed high purity. Scanning electron microscopy (FESEM) showed the spherical shape of the magnesium oxide particles. With particle size within the range (65-185) nm. While a (PMMA)-HAP nanocomposite was synthesized. -MgO) for use in dental applications such as fillings and dentures. A nanocomposite (PMMA-HAP-MgO) was manufactured by manual molding method by strengthening the poly methyl methacrylate polymer with certain weight ratios (1%,5%, and 10%) from a mixture of nano powder where the added ratios were (99%PMMA-1%HAP,95%PMMA-7.4%HAP-0.3%MgO, and 90%PMMA-9.4%HAP-0.3%MgO). The topographical properties (surface roughness) of the nanocomposite samples were studied by testing them with an atomic force microscope. The results showed an increase in the value of pure PMMA, where the use of nanoparticle filler by 5% (4.7% HAP - 0.3% MgO) affected both the roughness and the root mean square ratio of the distribution of nanoparticles on the surface of the composite and the shape of the particles led to a roughness more than the polymer pure. The antibacterial activity of the polymeric overlay was examined on the bacteria that cause dental caries for (Streptococcus mutans), where the zone of inhibition were (1 mm, 2 mm, 5 mm) for the nanocomposite and no activity for pure PMMA.

KEYWORDS: AFM, Antibacterial activity, FESEM, Nano-composite, XRD.

الخلاصة

يركز يركز العمل الحالي على تحضير جسيمات أوكسيد المغنيسيوم النانوية باستخدام املاح كلوريدات المغنيسيوم المائية ومسحوق هيدروكسيد الصوديوم بطريقة الترسيب كيميائية بسيطة وعند درجة حرارة التلدين ٧٠٠ سيليزية لمدة ٦ ساعات. تم فحص الخصائص الهيكلية والمورفولوجية لجسيمات أوكسيد المغنيسيوم النانوية باستخدام حيود الأشعة السينية (XRD) اظهرت مكعب متعدد البلورات وباستخدام معادلة شيرير أظهر حجم بلوري ١٧,٢٣ نانومتر، وأظهرت تحليلات EDS نقاء عالي، كما بين اختبار المجهر الإلكتروني الماسح للانبعاثات (FESEM) الشكل الكروي لجسيمات اوكسيد المغنيسيوم مع حجم جسيمات ضمن النطاق (٦٥-١٨٥) نانومتر. بينما تم تصنيع مركب نانو (MgO- -HAP). (PMMA)) لاستخدامه في تطبيقات طب الأسنان مثل الحشوات وصناعة الأسنان. تم تصنيع متراكب نانوي (PMMA-HAP-MgO) بطريقة التشكيل اليدوي عن طريق تقوية بوليمر بولي ميثيل ميثاكريلات بنسب وزنية معينة (من خليط مسحوق النانو (HAP-MgO). تمت دراسة الخصائص الطبوغرافية (خشونة السطح) لعينات المتراكب النانوي باختبارها بمجهر القوة الذرية وأظهرت النتائج زيادة في قيمة PMMA النقي، حيث ان استخدام حشو الجسيمات النانوية بنسبة ٥٪ (٤,٧٪ HAP -0.3 ٪) MgO) أثر في كل من الخشونة والجذر متوسط المربع نسبة التوزيع الجسيمات النانوية على سطح المركب وشكل الجسيمات ادى الى خشونة أكثر من البوليمر النقي. وتم فحص النشاط المضاد للبكتيريا للمتراكب البوليمري على البكتريا المسببة لتسوس الأسنان، حيث كانت قيم منطقة التثبيط لبكتريا (العقدية الطافرة) هي (١ مم، ٢ مم، ٥ مم) للمتراكب النانوي ولم يحدث اي نشاط لـ PMMA النقي.

INTRODUCTION

Each Artificial tooth need to be long – time biocompatible and withstand oral conditions. One of the most common tooth diseases is dental caries, the cavities develop if the decay has not been prevented it's around 80% of people in developed countries experienced dental decay caused by bacterial infection Streptococcus Mutans (S. Mutans). The microorganisms that cause tooth decay called Cariogenic bacteria. It can be passed on from a mother or a caregiver to youngsters [1]. Tooth decay usually begins on the chewing surfaces or the teeth's proximal contacts S. Mutans is added to the mix mostly from another person's mouth The germs infect the mouth and inhabit it as well as plaque development, cling to the teeth Plaque is a soft, sticky substance that forms on the surface of the body Food deposits a sticky coating on the teeth deterioration. The biofilm is hospitable to the spread of germs and bursting with microorganisms the bacteria S. Mutans decompose. Lactic acids are produced as a result of the fermentation of carbohydrates.

This is a process that causes tooth decay demineralization, or the loss of minerals calcium phosphate is a mineral that comes from the teeth

Structure The tooth "softens" as a result and finally collapses in on itself, leaving a cavity. Tooth rotting is a common problem [2]. As for children caries the high levels of S. Mutans which cause enamel damage, excessive sucrose consumption (candy Consumption) [3, 4]. Metal oxide nanoparticles have many advantages and are used in medical research and scientific studies due to their interesting properties and benefits over bulk materials [5]. Recent research has indicated that nano-oxide may improve the organic polymer's physical and optical characteristics as well as give resistance to cracking and aging brought on by environmental stress. Magnesium oxide nanoparticles have received a lot of interest. Due to its qualities including a high melting point and high purity, antibacterial properties against a variety of disease, nontoxicity, and stability so it was used in biomedical applications [6]. Dental restoration can be classified as intracoronary that is placed in a prepared cavity in the crown or extra coronal when place the tooth restorations outside (around the tooth), restorations usually placed directly into the tooth cavity and composed of modularly material that sets and became solid rigid material that conserved by the tooth walls [7]. Composites are used to cover the dental filling to protect it from falling out or to hide the shape of the distorted tooth or protect a weak tooth from breaking. The most materials are not permanent and many and the materials requirements used in dental are biocompatible, permanent bonding with tissue or bone, suitable appearance, shows efficiency function, and ability of repair or regeneration of missing or damaged tissues [8].

The humans tooth bone structure is composed of Enamel, Dentin, and Dental pulp the first protective layer of the humans tooth is Enamel it is made up of 96 percent calcium apatite, which comes in two forms: hydroxyapatite (HAP) [$Ca_{10}(PO_4)6(OH)_2$] and Fluor apatite [$Ca_{10}(PO_4)6F_2$] Crown contains 68% of collagen consist of a layer of dentin which covers the pulp that represents the inner layer of the tooth and the cementum layer it's the outer layer of the tooth root and its surrounds the dentin pulp, so the dental composites have to simulate the natural teeth mechanical properties such as hardness, compression and roughness thus we used nanotechnology to improve the dental. Composites involved metals, ceramics and polymers applied in teeth, but are still there some concerns of both chemical and physical and biological properties. Reviews in the terminal years pointed out that nanoparticles can be used to develop the mechanical properties of those materials [9, 10] [11]. Artificial teeth should closely resemble genuine teeth in terms of appearance and anatomical structure. Additionally, it should be nontoxic, not affect oral soft tissues, insoluble in saliva, and lightweight. Polymers have been used to make artificial teeth for persons who are missing teeth and need artificial restoration [12, 13], like Poly methyl Methacrylate (PMMA) resin, which is frequently used in dental laboratory to fabricate and repair orthodontic retainers and to reline prostheses and permanent crowns in dental clinics and industry (like fabrication of artificial teeth). PMMA has many benefits for dental applications, such as the capacity to tolerate high temperatures, ease of laboratory manipulation, lightness, low cost, stability of the oral environment, esthetic appeal, color, and lack of toxicity or mutagenicity to the oral tissues [14, 15, 16]. This research aimed to synthesize nano–composite (PMMA/HAP/MgO) by hand molding and

studying the structural, morphological properties, and antibacterial activity against (S. Mutans).

MATERIALS AND METHODS

Synthesis MgO NPs

Preparation of magnesium oxide by the chemical method, 0.2 mol of magnesium chloride hex hydrate (MgCl2.6H2O) was liquefied in 50 ml of pure water, and 0.4 mol sodium hydroxide powder in 25 milliliters of pure water, mix the two solutions using a burette drop by drop to obtain a white precipitate, then isolate the fluid mixture by centrifuge at 3000 rpm for 10 minutes to obtain a milky white precipitate, then wash the white precipitate with distilled water and ethanol twice at a time. respectively, to get rid of impurities and then dried at 100 °C and then calcined at 700 °C for 6 hours, to obtain white powder of MgO.

Synthesis of PMMA/HAP/MgO composite mold sample

To prepare PMMA/HAP/MgO composite mold sample it has been used corrosion-resistant steel with dimensions of 250×250×150 mm^3. the specimens were made corresponding to the guidelines for using heat cure PMMA polymer with a 2:1 weight ratio of powder to (glycoldimethacrylate) liquid. The powders (HAP/MgO) were mixed with by a magnetic stirrer for 15 minutes and mill grounded to obtain homogeneity. The liquid was combined to the powder in a 2:1 ratio and allowed until it reaches the dough stage. To eliminate any air bubbles in the dough, the mold is moved from the sides and the dough is placed in it. The mold is then tightly sealed and placed in an oven at 80 o C for 3 hours to finish the solidification process.

Antimicrobial Activity

The antimicrobial activity of the samples has been examined against bacterial cultures Gram-positive (Streptococcus mutans), were provided by the Microbiology laboratory of the University of AL-Mustansiriyah-College of science. This test carried out by method [17]. Nutrient agar was prepared by dissolving 3.8g of MUELLER-HINTON AGAR in100Ml of distilled water in a flask that boiled using Benzen burner. The flask is covered and placed in an autoclave for 15 minutes and at 15 bar pressure and 121°C to ensure a typical mixing after that the viscous solution in the flask and attached to cotton wool and steam sterilized at 121°C, 15 bar for 15 minutes. Then left to cool at 45 °C then poured into a 90 mm diameter sterile Petri dishes which are immediately covered and allowed to set for 30 minutes to solidify. By using inoculating loop 0.1 mL of bacteria and fungus suspension were spread on the solidified agar medium. After that, a 6 mm agar well was made with the help of Sterilized cork borer to make 5 Wells of 6 mm diameter. Then, a portion of each sample was introduced into the agar well. Subsequently, the growth was incubated at 37°C for 18-24 hours for bacteria. Following this, the diameter of the inhibition zone (in mm) surrounding the agar well, indicating antimicrobial activity, was measured.

RESULTS AND DISCUSSION

Structure and morphology of Nano MgO

X-ray diffraction (XRD) studies

Figure 1 Represents the relationship between intensity and ($2\boldsymbol{\theta}$) in range 27◦ and 80◦. The Nano material is polycrystalline and face center cube shaped. It obviously exhibits the peaks were absorbed 27.51◦, 31.83◦, 37.09◦, 43.07◦, 45.57◦, 56.5 6◦, 75.08◦, 78.77◦, and 84.03◦ along with Miller's indices value (300), (220), (111), (200), (400), (422), (311), (222), and (444) respectively. (JCPD No. 45.0946). Calculations obtained due to Scherer equation:

$$\text{Crystallite size} = 0.9\lambda/(\beta \cos\theta) \quad (1)$$

Where λ=1.54 Å is the wavelength of X-ray, FHWM is the breading of the diffracted peak at half maximum and θ is the Bragg angle. magnesium oxide diffraction peaks well indexed to poly crystalline cubic phase of magnesium oxide mentioned in the JCPDS database (No. 45.0946) The crystallite size for the strongest peak was 17.23 nm compared to the research[18, 19], which confirm that MgO NP was absence from any impurities. Lattice constant was a = 4.19 Å.

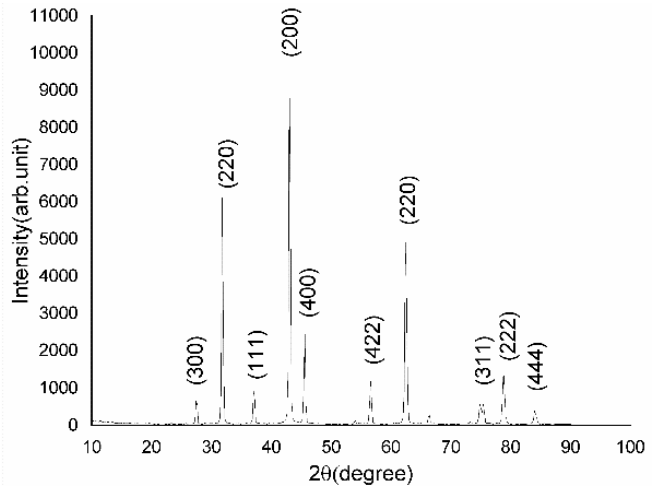


Figure 1. XRD pattern of MgO nanoparticles.

Field Emission Scanning Electron Microscopy studies (FESEM)

Figure 2 represented the surface structural and morphological properties of magnesium oxide nano-powder which exhibited nano-spherical forms that were well-uniformly crystalline and porous, with agglomeration occurring [20]. The particle sizes of Magnesium oxide samples fall in the range of (65-185) nm.

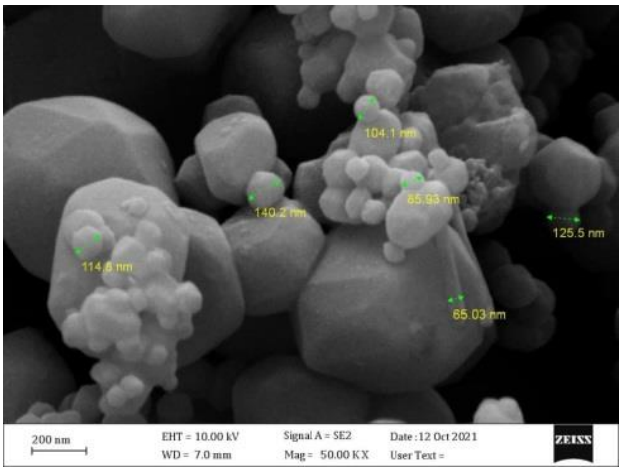


Figure 2. FESEM image of MgO NP.

Energy Dispersive Spectroscopy (EDS)

Figure 3 showed a quantitative analysis of MgO NPs was done by EDS, as there is an obvious strong peak comprising only of Mg and O, and there is another peak in the EDS where Magnesium is 58.2 % and the Oxide is 32.7%, this indicates that the MgO NPs are quite pure.

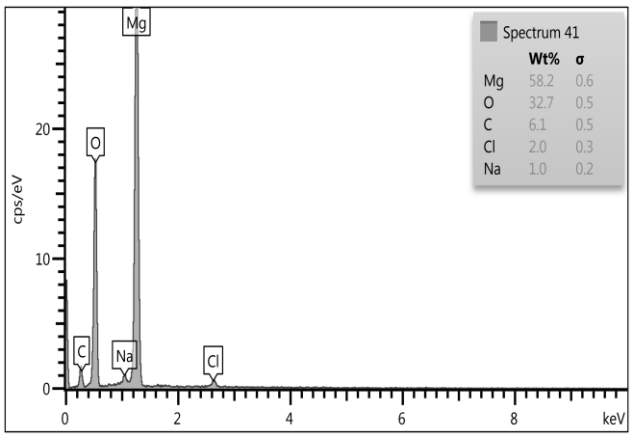
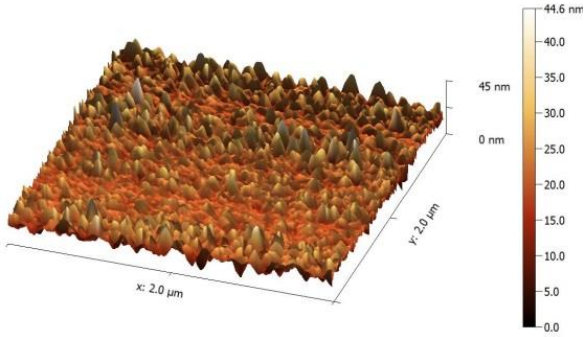


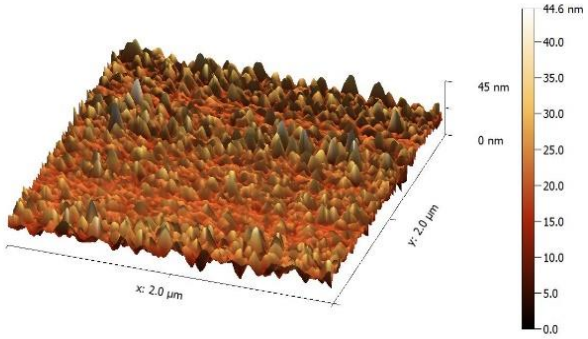
Figure 3. EDS analysis.

Atomic Force Microscopy (AFM) analyses for PMMA-HAP-MgO nano-composite

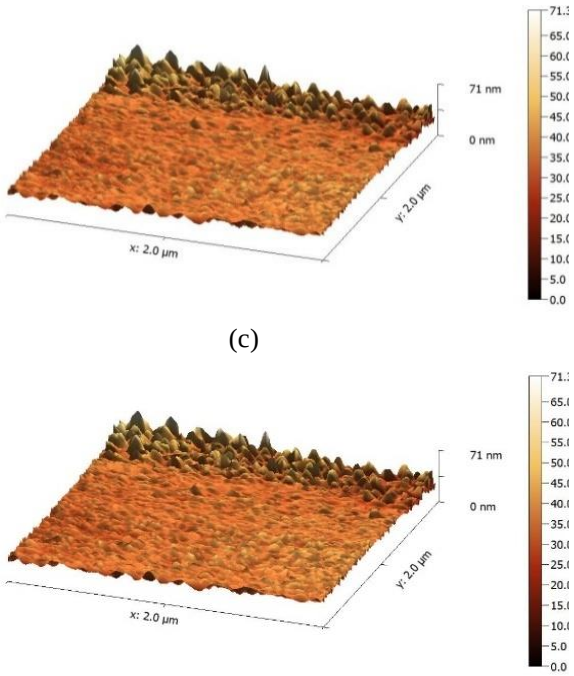
Figure 4 elucidate the AFM test in 3D and 2D images scan region 0.2µm *2.0µm of PMMA-HAP-MgO nano composite. The results of particle size distribution for these Nano composite of PMMA samples of different surface roughness displayed in Table 1. The composite with 5% nanoparticles filler (4.7%HAP - 0.3%MgO) as noted as MIX2 exhibits an increasing in the value of pure PMMA in both roughness and root mean square, meaning this filler ratio acquired the composite distribution of surface height and particles shape more than the pure polymer that led to an increase in roughness because the particles were produced by packing many nanocrystals [21].



(a)



(b)



(c)

(d)

Figure 4. AFM surface morphology of PMMA/HAP/MgO nano composite IN 3D: (a) Pure PMMA, (b) 99% PMMA, 1%HAP, (c) 95%PMMA, 4.7%HAP, 0.3%MgO, (d) 90%PMMA, 9.4%HAP, 0.6%MgO.

Table 1. AFM results of PMMA/HAP/MgO.

Composite	Pure PMMA	1%	5%	10%
Roughness average (Ra)/nm	3.04	2.01	3.1	2.00
Root Mean Square (RMS)/nm	4.05	2.72	4.33	2.85
Average diameter/nm	19.07	16.15	33.91	18.15

Antibacterial activity of PMMA/HAP/MgO Nano composites test

The anti-bacterial activity of the polymeric nano composite has been studied in terms of inhibition zones (mm). The inhibition zone has been measured against stander isolate bacteria (Streptococcus Mutans). Figure 5 showed the inhibition zone of nano composite (PMMA/HAP/MgO).

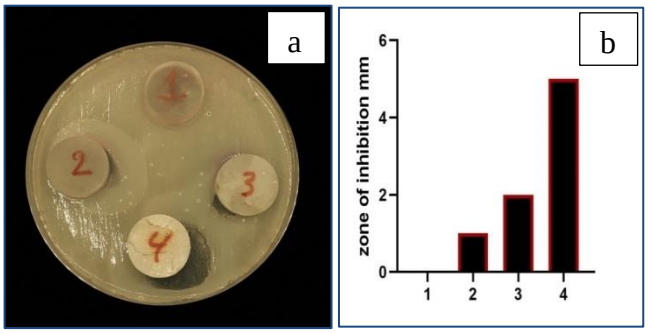


Figure 5. (a) Inhibition zone for nano composite (PMMA/HAP/MgO) samples against Streptococcus Mutans, (b) Statistical analysis.

The nano composite PMMA/HAP/MgO also showed high activity performance against S. Mutans with the increasing of HAP/MgO NPs. the inhibition zone values were (1mm,2mm,5mm) for the nanocomposite and zero activity for pure PMMA which indicate that there is no antibacterial activity for the pure PMMA polymer [22].

CONCLUSIONS

In this paper, magnesium oxide was prepared by simple chemical method (co-precipitation) and characterized by XRD, EDS and FESEM analysis. The crystallite size was calculated as 17.23 nm by XRD analysis, EDS analysis showed high purity, and from FESEM test particle size in the rang (65-185) nm, Antibacterial effective against S. Mutans increased with adding HAP/MgO NPs, AFM test showed an increase in the value of pure PMMA in both roughness and root mean square at nano-composite with 5%wt. (4.7%HAP-0.3%MgO) wt. nanoparticles filler sample.

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REFERENCES

[1] H. Çolak, Ç. T. Dülgergil, M. Dalli, and M. M. Hamidi, "Early childhood caries update: A review of causes, diagnoses, and treatments," Journal of natural science, biology, and medicine, vol. 4, no. 1, p. 29, 2013. https://doi.org/10.4103/0976-9668.107257

[2] Y. Zhang, X. Wang, H. Li, C. Ni, Z. Du, and F. Yan, "Human oral microbiota and its modulation for oral health," Biomedicine & Pharmacotherapy, vol. 99, pp. 883-893, 2018.

https://doi.org/10.1016/j.biopha.2018.01.146
[3] R. Smith, V. M. Badner, D. E. Morse, and K. Freeman, "Maternal risk indicators for childhood caries in an inner city population," Community dentistry and oral epidemiology, vol. 30, no. 3, pp. 176-181, 2002. https://doi.org/10.1034/j.1600-0528.2002.300303.x
[4] L. Haghshenas, A. Amini, A. B. Bhatti, and G. Rahimi, "In vitro antibacterial biofilm effect of magnesium oxide nanoparticles on Streptococcus mutans," Micro and Nano Biomedicine, pp. 21-27, 2016. https://doi.org/10.15412/J.MNB.05010104
[5] H. Fahmy et al., "Review on MgO nanoparticles nultifunctional role in the biomedical field: Properties and applications," Nanomedicine Journal, vol. 9, no. 1, pp. 1-14, 2022.
[6] B. A. A. ALwash, A. P. I. N. Yaseen, and A. P. A. A. L. Rashid, "Shear Bond Strength of Acrylic Denture Teeth to Different Pmma/Mgo% Reinforcing Nanofillers," MUSTANSIRIA DENTAL JOURNAL, vol. 17, no. 1, 2021.
[7] T. Nejatian, A. Johnson, and R. Van Noort, "Reinforcement of denture base resin," in Advances in Science and Technology, 2006, vol. 49: Trans Tech Publ, pp. 124-129. https://doi.org/10.4028/3-908158-05-2.124
[8] H. Reza Rezaie et al., "Dental restorative materials," A Review on Dental Materials, pp. 47-171, 2020. https://doi.org/10.1007/978-3-030-48931-1_3
[9] H. Xu et al., "Indentation damage and mechanical properties of human enamel and dentin," Journal of dental research, vol. 77, no. 3, pp. 472-480, 1998. https://doi.org/10.1177/00220345980770030601
[10] J. Nicholson and B. Czarnecka, Materials for the direct restoration of teeth. Woodhead Publishing, 2016.
[11] S. T. Ozak and P. Ozkan, "Nanotechnology and dentistry," European journal of dentistry, vol. 7, no. 01, pp. 145-151, 2013.
[12] M. H. Mohamed, "EFFECT OF DIFFERENT ARTIFICIAL TEETH MATERIALS ON THE SUPPORTING STRUCTURE OF IMPLANT RETAINED MANDIBULAR OVERDENTURE," Ain Shams Dental Journal, vol. 21, no. 1, pp. 135-143, 2021. https://doi.org/10.21608/asdj.2021.94476.1072
[13] D. B. Barbosa, V. A. R. Barão, D. R. Monteiro, M. A. Compagnoni, and J. Marra, "Bond strength of denture teeth to acrylic resin: effect of thermocycling and polymerisation methods," Gerodontology, vol. 25, no. 4, pp. 237-244, 2008. https://doi.org/10.1111/j.1741-2358.2008.00218.x
[14] G. Carlsson and R. Omar, "The future of complete dentures in oral rehabilitation. A critical review," Journal of Oral Rehabilitation, vol. 37, no. 2, pp. 143-156, 2010. https://doi.org/10.1111/j.1365-2842.2009.02039.x
[15] T. R. Meng and M. A. Latta, "Physical properties of four acrylic denture base resins," J Contemp Dent Pract, vol. 6, no. 4, pp. 93-100, 2005. https://doi.org/10.5005/jcdp-6-4-93
[16] M. S. Zafar, "Prosthodontic applications of polymethyl methacrylate (PMMA): an update," Polymers, vol. 12, no. 10, p. 2299, 2020. https://doi.org/10.3390/polym12102299
[17] R. Cruickshank, J. Duguid, B. Marmion, and R. Swain, "Test for sensitivity to antimicrobial agents," Medical microbiology, vol. 2, pp. 196-197, 1975.
[18] M. Sundrarajan, J. Suresh, and R. R. Gandhi, "A comparative study on antibacterial properties of MgO nanoparticles prepared under different calcination temperature," Digest journal of nanomaterials and biostructures, vol. 7, no. 3, pp. 983-989, 2012.
[19] J. Suresh, R. G. R. Gandhi, S. Selvam, and M. Sundrarajan, "Synthesis of magnesium oxide nanoparticles by wet chemical method and its antibacterial activity," in Advanced Materials Research, 2013, vol. 678: Trans Tech Publ, pp. 297-300. https://doi.org/10.4028/www.scientific.net/AMR.678.297
[20] K. Kaviyarasu, C. M. Magdalane, K. Anand, E. Manikandan, and M. Maaza, "Synthesis and characterization studies of MgO: CuO nanocrystals by wet-chemical method," Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy, vol. 142, pp. 405-409, 2015. https://doi.org/10.1016/j.saa.2015.01.111
[21] V. Pokropivny, R. Lohmus, I. Hussainova, A. Pokropivny, and S. Vlassov, Introduction to nanomaterials and nanotechnology. Tartu University Press Ukraine, 2007.
[22] A. H. Phakatkar et al., "Novel PMMA bone cement nanocomposites containing magnesium phosphate nanosheets and hydroxyapatite nanofibers," Materials Science and Engineering: C, vol. 109, p. 110497, 2020. https://doi.org/10.1016/j.msec.2019.110497

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