271 Egypt. J. Chem. **Vol. 66,** No. 6 pp. 197 - 203 (2023)

Egyptian Journal of Chemistry

http://ejchem.journals.ekb.eg/

Impact of embedded sol-gel synthesized triple composites on polymer's mechanical properties

Hayder Abbas Sallal[1], Mohammed S. Radhi[2], Mohammed Hamid Mahboba[3], Zainab Al-Khafaji [4]

1 Prosthetics and Orthotics Engineering Department, College of Engineering, University of Al-Nahrain, Jadriya, Baghdad, Iraq.

2 Ceramic and Building Materials Department, College of Materials Engineering, University of Babylon, Babylon, Iraq.

3 Technical Institute of Kufa - Al-Furat Al-Awsat Technical University, Iraq

4 Building and Construction Engineering Technology Department, AL-Mustaqbal University College, Hillah 51001, Iraq.

Abstract

The current study aims to prepare a nano-hybrid composite material reinforced with perlon fibers and triple nanoparticles (TiO2-ZnO-ZrO2) produced using the sol-gel method. The resulting sol-gel synthesized nanoparticles were evaluated using assays (EDX, X-Ray, SEM, and PSA). The results of the investigation revealed that using a temperature of 650 oC instead of 850 oC reduces the size of the particles. The other side included adding these nanoparticles according to weight ratios (3, 6, 9 and 12%) with a fixed number of layers of perlon fibers in the form of a mat (6 layers) to a polymeric blend consisting of (10% methyl vinyl silicon + 90% lamination) resin. The selected composite material was evaluated by tensile, flexural, impact, and hardness assays. The addition of the powder affected the mechanical behavior of the nano-hybrid composite material reinforced with the triple nanopowder, especially at the particle size (87 nm) and at the percentage (9% wt.), where the results of (tensile, bending, impact, and hardness) increased (83 MPa, 102 MPa, 13.8 kJ/m², 99 shore D) respectively, compared to the powder-free reference sample where the tensile, bending, impact, and hardness test values were (53 MPa, 85 MPa, 10.8 kJ/m², 83 shore D) respectively. This hybrid nanomaterial can be used in prosthetic applications such as the foot.

Keywords: nano-hybrid composite material, sol-gel, triple nanoparticles, (10% methyl vinyl silicon + 90% lamination) blend, mechanical behavior.

1. Introduction

The discovery of polymers helped a qualitative leap in the materials used in human life, as they were a successful alternative to many materials, in addition to their entry into many new uses that the rest of the engineering materials were unable to provide [1,2]. Polymers prompted many researchers to develop new materials, and this helped to find a new type of material, which is composite materials [3,4]. Composite materials have opened new horizons for scientific research because they are constantly updated and developed to support modern industries and provide ease of manufacture [5,6]. Modern nanomaterials have entered a great deal in the field of manufacturing composite materials and represented a wonderful duo with polymers in the development of the nanocomposite materials industry [7,8]. Nanoparticles were among the most widely used materials in all industrial and research fields because of their outstanding ability to develop and improve other materials if nanoparticles were added to them [9]. One of the most important types of particles used in this field is the oxides because of their high strength , hardness and chemical inertness [10,11]. The oxides of titanium, zirconia, zinc and aluminium are among the most important types of oxides used in many engineering, medical, civil, aerospace, and other applications because of their excellent mechanical properties and good medical, insulation, and chemical properties [12–14].

Many researchers studied the properties of composite materials and the effect of additives on the properties of polymers. Muhsin et.al, studied lamination resin, and a group of successive perlon and kevlar layers was used with a total of 7 layers to study their effect on giving better properties to the manufactured

*Corresponding author email: zainabcivil90@gmail.com; (Zainab Al-Kafaji).
Receive Date: 06 August 2022, Revise Date: 04 September 2022, Accept Date: 05 September 2022
DOI: 10.21608/EJCHEM.2022.154630.6684

socket for use in prosthetic limbs. The work included two parts: experimental and numerical. The work was based on studying the tensile property and ground reaction force. Whereas the experimental and numerical results showed that the socket made of the composite material resulting from the succession of layers was stronger than the polypropylene used for comparison in the research [15].

Also, lamination resin was used with several layers of successive (perlon, carbon and glass) fibres in different sequences to study their effect on giving better properties to the vacuum-formed socket for use in prosthetic limbs. The work included an experimental part to study the tensile and fatigue properties. Where the results showed that the composite material resulting from the use of two layers of carbon fibres with layers of perlon was the strongest type of reinforcement compared with other types of fibre used in the research [16]. Abdulaziz et.al studied used PMMA as the base material and nano (zirconia and titania), and glass fibres with different percentages of weight % were used as reinforcing materials to evaluate the bending resistance and hardness of the manufactured composite material. Through the results, it was shown that the flexural strength and hardness increased with the addition of nanoparticles. The composite materials that contain more than 3% of the filler materials are of high hardness [17].

Ameer et.al studied the lamination resin used and it was reinforced with multiple layers of carbon and perlon ranging from (10 to 12) layers in a different sequence, and then palm nut powder was added. The resulting composite material was subjected to tensile, bending and fatigue tests to know the effect of reinforcing materials on the properties of lamination. Through the results, the powder did not significantly improve the properties, and the best results were when using 3 layers of carbon fibres without powder [18]. Wei-li Wu studied the strengthening of Methyl vinyl silicon by dough-moulding compound (DMC) and silica was added to the mixture as a filler, in addition to the effect of these materials and curing conditions on the properties of the resulting composites. The best result was when adding (70 DMC and 35 silica) at a temperature of 170 degrees Celsius, where the mechanical properties such as hardness and tensile were improved in addition to that it gave a high bonding through FTIR test [19]. The objective of this work is to prepare triple compound nanoparticles and add them to the polymeric blend in addition to a fixed number of perlon layers (6 layers) to produce a hybrid nanocomposite material used in the applications of prosthetics.

2. Experimental.

The materials used in this research are titanium tetrachloride from Sigma-Aldrich Company (USA) and zinc chloride, and zirconium chloride from Thomas Baker India. As for the polymers, lamination resin from Otto Book, Germany, and methyl vinyl silicone resin from Polybit Company UAE.

A set of devices were used to examine and evaluate the powder and the nanocomposite material, as follows:

1-X-ray machine model (X-ray 6000: type Cu, NF) Shimadzu Company.

2- Scanning Electron Microscope and EDX (type Tescan), Vega Company.

3- Particle size analyzer (Nanoprook 90 Plus type) the USA.

4- Tensile and bending tests by Universal mechanical test device (Instron 1195).

5- Hardness device (type Shore D hardness) China.

6-impact test (type Izod machine) Germany.

Preparation of the composite ternary nanopowder (TiO_2-ZnO-ZrO_2). 1 molar of each of (titanium tetrachloride, zinc chloride, and zirconium chloride) is dissolved in distilled water separately, the following ratio is mixed (40: 40: 20) % of each of (zinc chloride: zirconium chloride: titanium tetrachloride) dissolved in water distilled and mix for 90 minutes after that surfactant (sucrose) is added and mixing continues for another 30 minutes, then drops of ammonium hydroxide are added to form a gel. After that, the filtered water is removed from the top of the container containing the gel and then filtered on filter paper to remove the remaining water. After the filtering process is completed, the remaining material is dried at a temperature of 115 °C for 4 hours, and the dried material goes to the calcination process at a temperature of (650 and 800) °C for 3 hours. The resulting powder is examined and evaluated by X-ray and EDX chemical composition examination, in addition to the particle size analysis and SEM structure examination.

The polymeric composite material will be manufactured by incorporating the matrix material, which is the polymeric blend (10% methyl vinyl silicone and 90% lamination) (which was selected according to the best mechanical properties among the various mixing ratios that were tested in the laboratory, where this ratio was the best when the properties were compared with the rest of the mixing ratios), with the strengthening material, which is represented by the triple compound nanoparticles and according to the weight fractions (3, 6, 9 and 12) %wt., in addition to the 6 layers of Prlon fibres. The samples were cut according to the ASTM. The hybrid nanocomposite material will be evaluated by its mechanical behaviour using tests (tensile, bending, impact, and hardness).

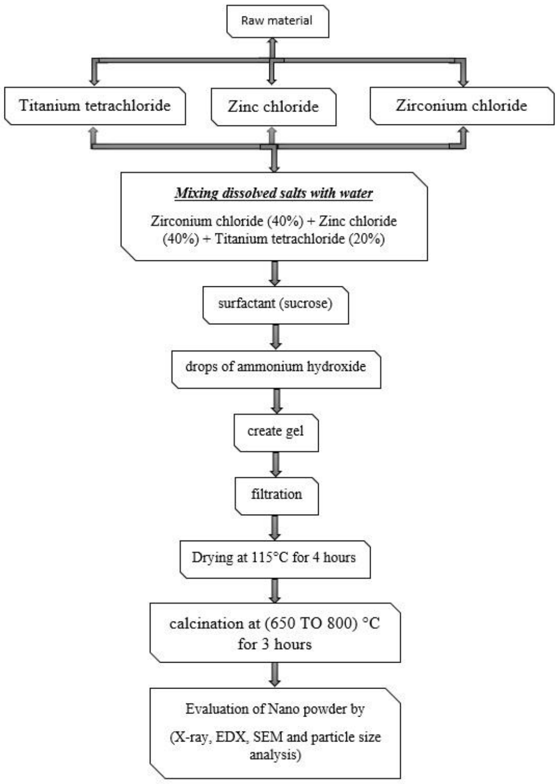


Fig. 1. Technical path for the preparation of the nanopowder.

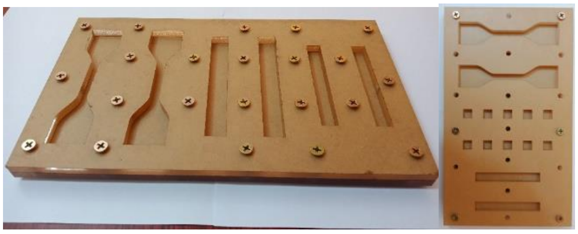


Fig. 2. Casting mould used for preparing samples of hybrid nanocomposite material.

3. Results and discussions.

3.1. Ternary Nano Powder tests.

3.1.1. Chemical analysis EDX test.

Fig.3, the composition of the ternary nanocomposite powder that the resulting powder is pure through the presence of the elements oxygen, titanium, zinc, and zirconium, and there are no impurities within the composition at both temperatures (650 and 800)ºC, and the presence of oxygen indicate the presence of oxides. This examination also indicates the type of material present, in addition to having other tests such as X-rays, we can know the type of the powder. This proves the purity of the materials produced in this way [20]. In many cases, the reason for the purity of the material is the use of a correct method of manufacture [21].

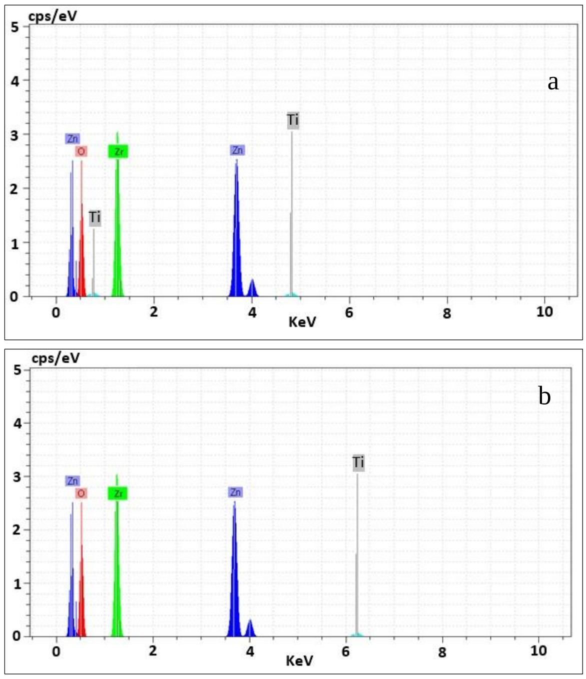


Fig. 3. EDX test: a) particles calcination at 650 ºC, b) particles calcination at 800ºC.

3.1.2. X-ray test.

The X-ray examination shown in fig. 4, shows the appearance of the anatase and rutile phases of titania at a temperature (650 **ºC**) according to JCPDS card no(21_1272) and (21_1276) respectively, in addition to that the addition of titania led to the appearance of two (m and t) phases of zirconia according to JCPDS card no (81_1314) and (81_1544) respectively [22,23]. therefore the chemical composition plays a role in the emergence of phases and reducing Calcination temperature, and the emergence of the phase of (anatase) for titania and phase (tetragonal) of zirconia at temperature (800 **ºC**), temperature has an effect on the formation of phases within the material [22,24].

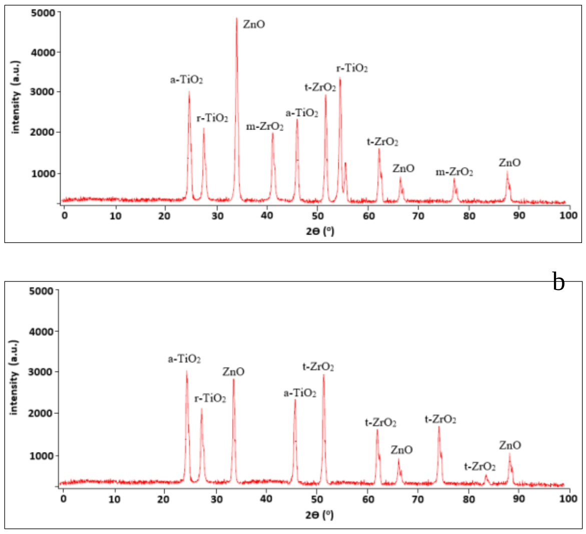
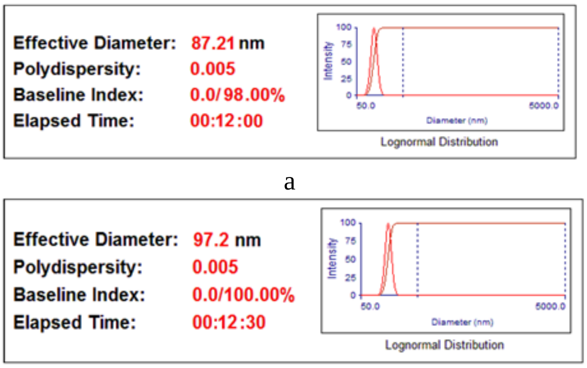


Fig. 4. X-Ray test: a) particles calcination at 650 ºC, b) particles calcination at 800ºC.

3.1.3. PSA test.

Fig. 5, shows the particle size of the ternary compound nanopowder resulting from the calcination process at two different temperatures. Where calcination at temperature (650 **°C**) gave grain size (87nm), while calcination at temperature (800 **°C**) gave grain size (97 nm). Therefore, this difference in granular size resulted from the difference in temperatures in addition to the chemical composition and the presence of titania may have helped in this [25,26]. There is no clear difference between the two sizes through the diagram because of the small difference in nanoscale between the two types, but the difference will be clear when there is a large difference between the sizes of the particles [27,28].



b

Fig. 5. PSA test: a) particles calcination at 650 °C, b) particles calcination at 800°C.

3.1.4. Scanning Electron Microscope (SEM) test.

The fig. 6. a) Represents the SEM image that gives an overview of the shape and the assembly of the particles inside the calcined ternary composite nanopowder at (650 **°C**) and the fig. 6. b) For the calcined nanopowder at (800 **°C**). Where notice through the pictures that the particles tend to be spherical than random, and we also notice the presence of some agglomeration of the particles because of the small size, which gives preference to the occurrence of clumps due to the small surface area of the particles [29,30].

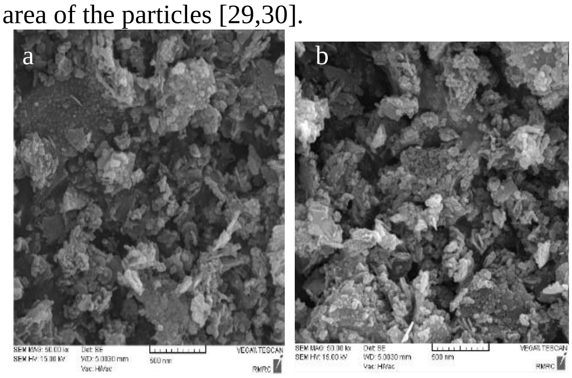


Fig. 6. SEM test: a) particles calcination at 650 °C, b) particles calcination at 800°C.

3.2. Mechanical test for hybrid nanocomposites reinforced by ternary composite Nano powder.

3.2.1. Tensile test.

Fig.7. gives an idea about the tensile properties. The curve represents the relationship between the weight ratios of the nanopowder and the resulting ultimate tensile strength of the hybrid composite material. From the figure, we notice that the maximum tensile strength has improved well when adding the triple compound powder, where the highest results were at the weight ratio (9%) for both particle sizes (87 and 97) nano. The mechanical properties of composite materials improve upon strengthening with materials of higher resistance (such as ceramic and metal materials) than the base material to achieve the principle of integrating properties to obtain the best and improved mechanical properties [31,32].

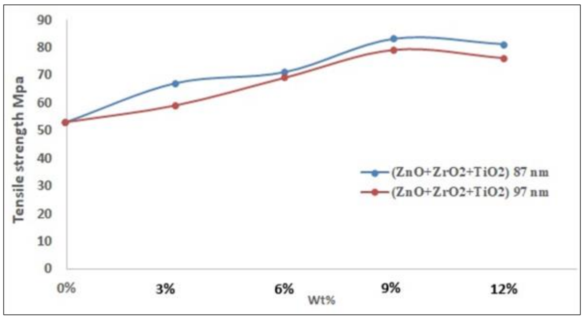


Fig.7. Relationship between tensile strength and weight fraction of ternary nanocomposite powder.

3.2.2. Bending test.

Fig. 8, shows the relationship between the weight ratios of the ternary composite nanopowder and the resulting bending strength of the hybrid composite material. From the figure, we notice that the bending resistance has improved well when adding nanoparticles to all weight ratios compared to the base sample, where the highest results were at the weight ratio (9%) for both granular sizes (87 and 97) nano. Increasing the bonding between the components of composite materials leads to an increase in the strength of the interface formed between the matrix material and the different reinforcement materials, thus obtaining the best improved mechanical properties [32,33].

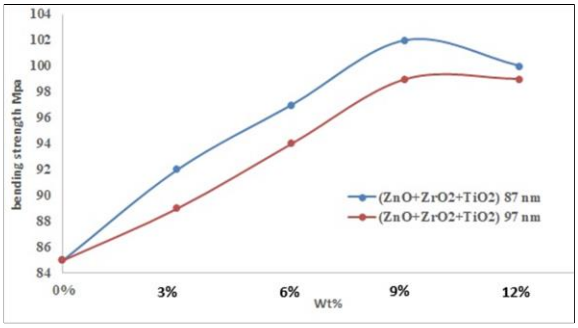


Fig. 8. Relationship between bending strength and weight fraction of ternary nanocomposite powder.

3.2.3. Impact test.

Through fig. 9, gives a picture of the impact resistance of the hybrid composite material. We note that the impact resistance has improved well when adding the compound triple nanoparticles to all weight ratios compared to the base sample, where the highest results were at the weight ratio (9%) for both granular sizes (87 and 97) nano. The greater the strength of the interface, the greater the resistance and durability of the material, and this is caused by the material used in the reinforcement, as ceramic materials are characterized by resistance and porosity, and these properties increase the strength of the brown surface and thus lead to improving the properties of the resulting materials [34,35]. The use of fibre enhances durability and helps dissipate absorbed energy [36].

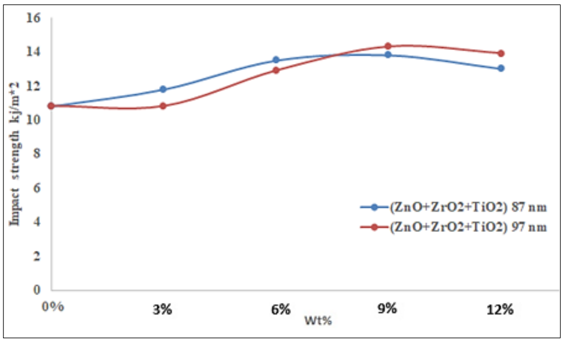


Fig. 9. Relationship between impact strength and weight fraction of ternary nanocomposite powder.

3.2.4. Hardness test.

Through fig. 10, gives a picture of the hardness of the hybrid composite material. We note that the hardness increased when adding the compound triple nanoparticles to all weight ratios compared to the base sample, where the highest results were at the weight ratio (12%) for both granular sizes (87 and 97) nano. Ceramic materials are characterized by high hardness compared to polymers, so when added, they cause an increase in the hardness of the polymer in addition to the size and shape of the added particles, it gives an advantage in increasing the hardness, as the smaller the granular size, the greater the dispersion inside the matrix [37]. In addition to the porosity of the ceramic materials, which leads to increased mechanical bonding with the matrix [38].

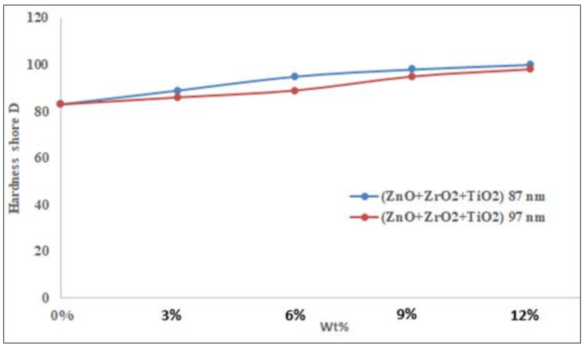


Fig. 10. Relationship between hardness shore D and weight fraction of ternary nanocomposite powder.

4. Conclusion.

After the appearance and study of the results, it was found that:

The triple composite nanopowder calcined at a temperature of (650°C) gave nanoparticles with less particle size and more phases compared to the powder calcined at a temperature (850°C).

The addition of the powder had an effect on the mechanical behaviour of the nano-hybrid composite material reinforced with the triple nanopowder, especially at the particle size (87 nm) and at the percentage (9%), where the results of (tensile, bending, impact and hardness) increased (83 MPa, 102 MPa, 13.8 kJ/m², 99 shore D) respectively, compared to the powder-free reference sample where the tensile, bending, impact and hardness test values were (53 MPa, 85 MPa, 10.8 kJ/m², 83 shore D) respectively.

From the results, we can choose the hybrid composite material for the applications of the prosthetics industry (Foot), which consists of a polymeric substrate (10% methyl vinyl silicon + 90% lamination) and a resin reinforced with (6 layers) perlon fibres with nanoparticles of size (87 nm) and by (9% wt.).

5. Acknowledgement

The Ministry of Higher Education, University of Babylon and Al-Mustaqbal University College is gratefully acknowledged. This research was carried out in the laboratory at University of Babylon.

6. Reference

[1] Borchardt JK. Blending provides benefits-Composites. Mater Today 2004;4:19.

[2] Abed Janabi ZM, Jaber Alsalami HS, Al-Khafaji ZS, Hussien SA. Increasing of the corrosion resistance by preparing the trivalent nickel complex. Egypt J Chem 2021.

[3] Vandeginste V. Food waste eggshell

valorization through development of new composites: A review. Sustain Mater Technol 2021;29:e00317.

[4] Ali Abdulkhabeer Ali, Ali Abdulhakeem Naghmash. Synthesis and evaluation of demulsifier in crude oil treatment. AIP Conference Proceedings 2443, 030040 (2022); https://doi.org/10.1063/5.0091943.

[5] Fakirov S. Composite materials—is the use of proper definitions important. Mater Today 2015;18:528–9.

[6] Bhattacharyya D, Fakirov S. Synthetic polymer-polymer composites. Carl Hanser Verlag GmbH Co KG; 2012.

[7] Wong LW, Goh CBS, Pasbakhsh P, Tan JBL. Natural hollow clay nanotubes and their applications as polymer nanocomposites in tissue engineering. J Sci Adv Mater Devices 2022:100431.

[8] Wei Y, Zhou H, Deng H, Ji W, Tian K, Ma Z, et al. "Toolbox" for the processing of functional polymer composites. Nano-Micro Lett 2022;14:1–41.

[9] Kraleva E, Saladino ML, Matassa R, Caponetti E, Enzo S, Spojakina A. Phase formation in mixed TiO2-ZrO2 oxides prepared by sol-gel method. J Struct Chem 2011;52:330–9.

[10] Nga NK, Chau NTT, Viet PH. Preparation and characterization of a chitosan/MgO composite for the effective removal of reactive blue 19 dye from aqueous solution. J Sci Adv Mater Devices 2020;5:65–72.

[11] Sallal HA, Mohammed MS, Kadhim IAU. The Nanotechnology (Preparation, Applications, and Cancer Treatment). Iraqi J Cancer Med Genet Vol 2020;13:22.

[12] Grigorie AC, Muntean C, Vlase T, Locovei C, Stefanescu M. ZnO-SiO2 based nanocomposites prepared by a modified sol-gel method. Mater Chem Phys 2017;186:399–406.

[13] Esposito S. "Traditional" sol-gel chemistry as a powerful tool for the preparation of supported metal and metal oxide catalysts. Materials (Basel) 2019;12:668.

[14] AlSaidy HAH, Alhasan DAH, Ali AA. Reviews on Plants Gum and Mucilage I: The Composition and Pharmacological Significance of Turmeric (Curcuma longa) Rhizomes Polysaccharides and Mucilage/their Aqueous Extracts Commercial Products. Univ Thi-Qar J Sci 2021;8:14–25.

[15] Jweeg MJ, Ahumdany AA, Jawad AFM. Dynamic Stresses and Deformations Investigation of the Below Knee Prosthesis using CT-Scan Modeling. Int J Mech Mechatronics Eng 2019;19.

[16] Abbas SM. Effects of composite material layers on the mechanical properties for partial foot prosthetic socket. Al-Nahrain J Eng Sci 2018;21:253–8.

[17] Radhi NS, Sahi NM, Al-Khafaji Z. Investigation Mechanical and Biological Properties of Hybrid PMMA Composite Materials as Prosthesis Complete Denture. Egypt J Chem 2022.

[18] Jweeg MJ, Hamzah HA, Al-Waily M, Al-Shammari MA. A Finite Element Simulation of Nano Effects on Stress Distribution in a Below Knee Prosthetic. IOP Conf Ser Mater Sci Eng, vol. 1067, IOP Publishing; 2021, p. 12141.

[19] Wu W. Dough-moulding compound reinforced methyl vinyl silicone rubber with silica filler. Polym Polym Compos 2011;19:677–80.

[20] Haider AJ, Jameel LZN, Taha SY. Synthesis of TiO2 nanoparticles by using sol-gel method and its applications as antibacterial agents. Eng Tech J 2014;32:418–26.

[21] Mansoor A, Khan MT, Mehmood M, Khurshid Z, Ali MI, Jamal A. Synthesis and characterization of titanium oxide nanoparticles with a novel biogenic process for dental application. Nanomaterials 2022;12:1078.

[22] Shao GN, Imran SM, Jeon SJ, Engole M, Abbas N, Haider MS, et al. Sol–gel synthesis of photoactive zirconia–titania from metal salts and investigation of their photocatalytic properties in the photodegradation of methylene blue. Powder Technol 2014;258:99–109.

[23] Houšková V, Štengl V, Bakardjieva S, Murafa N. Photoactive materials prepared by homogeneous hydrolysis with thioacetamide: Part 2—TiO2/ZnO nanocomposites. J Phys Chem Solids 2008;69:1623–31.

[24] Mirković M, Filipović S, Kalijadis A, Mašković P, Mašković J, Vlahović B, et al. Hydroxyapatite/TiO2 Nanomaterial with Defined Microstructural and Good Antimicrobial Properties. Antibiotics 2022;11:592.

[25] Bessekhouad Y, Robert D, Weber J V. Preparation of TiO2 nanoparticles by Sol-Gel route. Int J Photoenergy 2003;5:153–8.

[26] Sallal HA, Abdul-Hamead AA, Othman FM. Effect of nano powder (Al2O3-CaO) addition on the mechanical properties of the polymer blend matrix composite. Def Technol 2020;16:425–31.

[27] Álvarez-Chimal R, García-Pérez VI, Álvarez-

Pérez MA, Tavera-Hernández R, Reyes-Carmona L, Martínez-Hernández M, et al. Influence of the particle size on the antibacterial activity of green synthesized zinc oxide nanoparticles using Dysphania ambrosioides extract, supported by molecular docking analysis. Arab J Chem 2022;15:103804.

[28] Bapat RA, Yang HJ, Chaubal T V, Dharmadhikari S, Abdulla AM, Arora S, et al. Review on synthesis, properties and multifarious therapeutic applications of nanostructured zirconia in dentistry. RSC Adv 2022;12:12773–93.

[29] Al-kaisy HA, Al-Gebory L. Preparation and Characterization of TiO2-ZrO2 Coating Prepared by Sol-Gel Spin Method. IRAQI J Mech Mater Eng 2020;20:257–67.

[30] Dubey RS, Rajesh Y, More MA. Synthesis and characterization of SiO2 nanoparticles via sol-gel method for industrial applications. Mater Today Proc 2015;2:3575–9.

[31] Núñez-Decap M, Wechsler-Pizarro A, Vidal-Vega M. Mechanical, physical, thermal and morphological properties of polypropylene composite materials developed with particles of peach and cherry stones. Sustain Mater Technol 2021;29:e00300.

[32] Mazur KE, Jakubowska P, Gaweł A, Kuciel S. Mechanical, thermal and hydrodegradation behavior of poly (3-hydroxybutyrate-co-3-hydroxyvalerate)(PHBV) composites with agricultural fibers as reinforcing fillers. Sustain Mater Technol 2022;31:e00390.

[33] Cunha F, Galante R, Bessa J, Vieira S, Nascimento J, Barroso H, et al. Mechanical properties and antimicrobial activity of pumice stone/sludge filled thermosetting composites. Sustain Mater Technol 2021;30:e00348.

[34] Islam MS, Masoodi R, Rostami H. The effect of nanoparticles percentage on mechanical behavior of silica-epoxy nanocomposites. J Nanosci 2013;2013.

[35] Sallal HA, Abdul-Hameed AA, Othman F. Preparation of Al2O3/MgO Nano-Composite Particles for Bio-Applications. Eng Technol J 2020;38:586–93.

[36] Mahboba MH, Sallal HA, Ali FH. Effect of particles size of zirconia on the mechanical behavior of the polymer blend. AIP Conf Proc, vol. 2213, AIP Publishing LLC; 2020, p. 20310.

[37] Won JS, Kwon M, Lee JE, Lee JM, Kwak TJ, Lee SG. Polyphenylene sulfide-modified carbon fiber reinforced high-strength composites via electrophoretic deposition. J Sci Adv Mater Devices 2022;7:100456.

[38] Wang Y, Zheng M, Liu X, Yue O, Wang X, Jiang H. Advanced collagen nanofibers-based functional bio-composites for high-value utilization of leather: A review. J Sci Adv Mater Devices 2021;6:153–66.