An Overview of Titanium Dioxide Effect on Mechanical Properties of PMMA-TiO2 Nanocomposites

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Abstract

Polymethyl methacrylate (PMMA) is a type of polymer that is currently widely used in dentistry due to aesthetic considerations, biocompatibility, availability, and ease of use. However, its mechanical strength is not sufficient for the mechanical requirements of the prosthesis.

Attempts have been made to increase its mechanical strength by integrating high mechanical performance TiO2 particles. This article comprehensively reviews the mechanical strength of PMMA reinforced by TiO2 nanoparticles.

Aspects of mechanical strength reviewed include tensile strength, compressive strength, creep strength, hardness, impact strength, and fracture toughness. The material's microstructure is discussed to strengthen the review of the mechanical strength.

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Introduction

Poly(methyl methacrylate) PMMA is a synthetic polymer derived from methyl methacrylate (MMA) ($C_5H_8O_2$) by a polymerization process, and it is often used as a denture material. It is a transparent and rigid thermoplastic. PMMA is regarded as a preferable material for a number of biomedical applications because of its high processability, handling characteristics, and biocompatibility.[1] In biomedical applications, especially for denture materials, this material replaces the previous denture material which is made of porcelain[2,3], gold[4,5], vulcanite, aluminum[2,3], and bakelite[2] which has many weaknesses. This replacement was carried out on the consideration of its satisfactory mechanical properties, ease of manufacturing process, good appearance, biocompatibility, low toxicity, and low cost.[6,7,8,9]

The mechanical properties of the PMMA are however not optimal, especially the strength and ductility, and leave a lot to be improved. Given that dentures are used primarily by the elderly, often with mobility constraints, there have been disruptions and a large issue with repairing fractured products.[10] Consequently, it is important to increase the mechanical characteristics of the denture material without compromising the existing advantages including aesthetic appeal and biocompatibility.

A study examined several changes in the chemical, reinforcement, and various cures to address the weakness of PMMA-based materials.[11] A study by Wang et al.[12] recently described the inclusion of nanoparticles in denture resins to enhance mechanical characteristics. Titanium dioxide, as it has extraordinary characteristics in both micro and nanoscale, plays an important role for researchers.[12,13]

Various works have been carried out in order to enhance the properties of PMMA by integrating particles into the host matrix.[14,15,16,17,18,19] A number of studies have reached the conclusion that the mechanical properties of heat-cured PMMA-based materials can be improved by using several types of particles introduced into the resin matrices.[20,21,22,17,18] This is a common method used to enhance composite mechanical characteristics. Among the particles that have been widely used for this strengthening function

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is Titanium dioxide (TiO2).[21,23,24,17,18]. TiO2 particles have been widely applied in the field of biomaterials, especially in dentistry because of their color similar to natural human teeth, and their high biocompatibility.[25,26] The TiO2 nanoparticles (NPs) also have a good modulus of elasticity (approximately 230 GPa). In addition, its white color, low toxicity, and high internal stability make TiO2 the main dental material.[27]

This review aims to provide information on the effect of the incorporation of TiO2 nanoparticles into the PMMA host matrix as a denture material. The articles reviewed include sources of data and information available from various sources including scientific articles, reviews, and published abstracts that review the mechanical properties of TiO2-PMMA composites including tensile and compressive strength, creep strength, hardness, impact strength, fracture toughness, and microstructural characteristics.

Mechanical Properties

A wide range of filler particles has been studied to enhance the characteristics of PMMA. Due to their high area and better distribution characteristics, the benefits of the use of nanoparticles can be obtained.[28,29] The effect of adding TiO2 to PMMA-based denture materials is explained in the following explanation which reviews the studies that have been carried out.

Particle effect on tensile properties

Various studies have been conducted to find out more about the effect of TiO2 on the PMMA matrix on the tensile strength of the material. It has been reported by Shirkavand and Moslehifard[30] that the tensile strength of composites increased up to 35% compared to pure PMMA by the addition of 1 wt.% TiO2. Further addition causes agglomeration of particles which are considered as defects and stress concentrations which can then act as crack precursors that reduce the tensile strength of the composite.[31,32] The study conducted by Shirkavand and Moslehifard showed that the composite with a TiO2 concentration of 1wt.% showed superior tensile properties compared to other compositions.[30] It is shown that the tensile strength, maximum stress, modulus of elasticity, and toughness modulus of the material increase with increasing particle content up to 1 wt.% and decreased with increasing particle content above 1wt.%. However, there are studies that found that at 15 wt.% TiO2 content increased the tensile modulus of composites to about 90% of pure PMMA.[26] This phenomenon is related to the strong adhesion at the particle-matrix interface so that the load is effectively transferred at the interface to the particles.

Many works have reported that the flexural strength evaluation is very important as the main mode fracture for the loading characteristics applied.[33,34] Many studies have shown that the flexural strength of resin is restricted and the introduction of TiO2 is not increased due to the cluster formation in the host acrylic matrix, which causes the composite to develop an unloadable area as a result of not being enveloped by the PMMA matrix.[35,36,37] In a study, Alhotan and co-workers prepared composites with acrylic resin as the matrix formed by conventional hot polymerization processes of PMMA powder which had added methyl methacrylate monomer as the liquid component. The TiO2 nanoparticles with weight concentrations of 1.5%, 3%, 5%, and 7% were added to acrylic. They reported that increasing the content of TiO2 nanoparticles in the PMMA resin decreased the flexural strength which gradually decreased by about 2 MPa with each increase in the TiO2 content added.[23] Nazirkar and co-workers attributed the presence of TiO2 NPs agglomerates, which act as impurities and plasticizers that interfere with the polymerization process, as the cause of the decrease in flexural strength.[21]

Numerous investigations also discovered that even after TiO2 particles were added to PMMA resin, the flexural strength kept rising. Increasing the interfacial bond strength between TiO2 and PMMA can improve the mechanical properties of the material, including flexural strength.[38,39,40] An increase in the value of flexural strength occurred when NP TiO2 content of 1wt.%, 2wt.%, and 5wt.% was added. It was also explained that this phenomenon occurs due to the reduction in the size of the filler, which binds the polymer matrix thereby increasing the flexural strength of the composite.[39] This argument is corroborated by other studies which confirmed that silanization of TiO2 NPs at a content of 1 wt.% increased the intermolecular bond strength, increasing the binding force at the interface between PMMA and TiO2 NPs.[41,42] In addition, the absence of agglomeration and good dispersion of TiO2 NPs are also the main reasons for the increase in flexural strength.[43]

In order to examine the mechanical properties of the produced nanocomposite with additional filler, Naji and colleagues developed an alkali hydrothermal process for TiO2 nano-tubes where TiO2 was incorporated in the PMMA.[44] The flexural strength was notably greater with a TiO2-nanotubes content of 2,5wt.% and 5wt.% than in the control specimens. The toughness of the fractures in produced filled samples has also been improved by up to (5%) NT-TiO2 and strengthened (except for the one filled with 2.5% Titania NTs).

An elasticity modulus at the surface of TiO2-PMMA nanocomposites has been measured by Alrahlah and colleagues by using a nanoindentation machine at room temperature.[45] It was reported that the addition of TiO2 nanoparticles at weight concentrations of 1, 2, and 3% resulted in a rise in the modulus of elasticity of 10%, 16%, and 29%, respectively, as measured at a penetration depth of 1200 nm.[45] The nano-modulus measured at various penetration depths tends to decrease Improvement of the mechanical properties of PMMA-based cement through the incorporation of n-TiO2 was also reported by Khaled et al.[46] which was later confirmed by another study conducted by Dafar and co-workers who reported that Young's modulus increased with reinforcement of n-TiO2 in the composite.[47]

Particle effect on compressive strength

Composite compressive strength is a material property that often colludes with other objects in its implementation.[48] The compressive force of PMMA filled with TiO2 treated with titanate was reported to be higher than those of TiO2 not treated.[49] TiO2 was also reported as a pigment in fixed prostheses composite opaques, which was treated with titanate.

In the cylindrical PMMA-NP TiO2 composite with a diameter of 25 mm and a length of 38 mm, the compressive strength did not show a significant difference in compressive strength between the control specimens and the three composites reinforced with TiO2 NPs which were varied by 0.5, 1 and 2wt.%.[50] The difference in compressive strength was also not significant between the control group and the composite reinforced with 2wt.% TiO2 NPs, although there was an increase in compressive strength from 23.04 MPa to 24.30 MPa.[51] A significant increase in compressive strength actually occurred in cold-cured PMMA, which increased by 3.3% from the compressive strength of the control specimen of 15.37 MPa.[51] The profile of compression versus deformation force shows that the largest deformation is in TiO2-reinforced heat cure PMMA, and vice versa, the smallest deformation is found in unreinforced cold cure PMMA.[51,52]

Particle effect on creep behavior

The creep behavior of TiO2-PMMA nanocomposites was observed using the RSA-G2 analyzer to evaluate their creep-recovery and relaxation behavior. The material creep behavior has been evaluated by creep testing at different loading and unloading of 10, 15, and 20 N over 240 minutes at 37°C. The RSA-G2, which is also used to investigate relaxation behavior, was also tested for 180 minutes under 1% strain.[45] They reported that the material was in the early stages deformed elastically in a really small space-time as a result of stress growing steadily at steady stresses. After unloading, the rest of the strain was seen as an ongoing deformation and a portion of the versatile strain quickly recovered while the rest of the cramped strain recuperated over time. After loading for 240 minutes, the creep strain reached 2.4% at an applied load of 20 N. This phenomenon is similar to what has been reported by other studies which reported that an increase in load caused an increase in the creep strain.[53,54]

The creep relaxation behavior is time-dependent, i.e., when the strain has not reached 1%. The phenomenon shown is the response to strain with a decrease in stress over time even though the stress is only about 30% of the initial value after more than 3 hours. Incorporating TiO2 nanoparticles leads to an increase in matrix stiffness by reducing the free-volume region and molecular chain movability of the PMMA matrix.[45]

Particle effect on hardness

Many studies show that the surface hardness of PMMA increases by the incorporation of TiO2 NP.[39,55] There are two factors associated with the increase in surface hardness properties of PMMA: the accuracy of the filler composition and the good interface bonded between the filler and the matrix.[56] This increase begins when the filler fraction reaches 1wt.% and at 2wt.% TiO2, the surface hardness value is the highest.[51] Regarding this composition, it can be explained that the matrix effectively covers the filler and high energy is required to break the matrix-particle bonds due to the high interfacial bond between the particles and matrix.[31] Incorporation of 1 to 3 wt% TiO2 NPs can increase the hardness up to 30% higher than pure PMMA.[57]

This finding is confirmed by a study conducted by Alrahlah et al.[45] Therefore, the most effective increase in PMMA hardness is the addition of 3% TiO2[55] although there are studies that suggest adding TiO2 NPs up to 5wt.% to increase the surface hardness even more, especially for conventional and high-impact heat cure PMMA.[38] This phenomenon is elucidated because the presence of TiO2 NPs in that content restricts the plastic deformation of the PMMA matrix.

Investigations on the hardness of TiO2-PMMA nanocomposites with variations in the weight content of TiO2 NPs of 1%, 2%, and 3% showed that at the same penetration depth, there was an increase in hardness of 18%, 24%, and 35%, respectively. Various values of nano hardness (H) were measured using formula (1) as follows.[45]

$$\boxed{H = 2.2\,(h)^{-0.35} + 0.2\,(wt.\%)} \quad (1)$$

where h is the penetration depth (in nm) and wt.% is 1, 2, and 3. Nano hardness as a function of penetration depth.[45]

In the 1990s, researchers found a tube type of hydrothermally synthesized TiO2 nanostructures. Of that type, linear nanotubes have a larger surface area. TiO2 nanotubes are one type of nanotubes that have relatively large specific surface areas of 170-250 m2/g.[58,59] The effects of the addition of titanium nanotubes (n-TiO2) to the surface hardness of polymethyl methacrylate (PMMA) used as a denture base have been investigated by Naji and colleagues.[44] They reported that the surface hardness of n-TiO2-PMMA nanocomposites with 2.5 and 5 wt.% of n-TiO2, was significantly higher than the control groups. They elucidate that the oriented nanotubes can aid in preserving reinforced polymer cohesion. Another relevant study reported that the increased hardness of PMMA composites reinforced with TiO2 nanotubes was due to the effect of the nanotubes being well dispersed in the matrix.[60] The length of the synthetic nanotubes used also plays a very large role in increasing the hardness of the composite considering the higher contact area at the nanotubes-polymer interface.[60,61] However, not much literature discusses the hardness of PMMA reinforced by TiO2 nanotubes. However, many studies report that TiO2 nanotubes have been successfully applied in biomedical fields such as bio-scaffolds and implant materials.[62,44]

Particle effect on impact strength

As a material for biomedical applications, PMMA-based materials are often subject to loading which causes deformation. For example, in the case of mastication and PMMA as a denture material, the effect of deformation is more of a concern than the impact strength of the material. Therefore, it is necessary to increase deformation resistance and stress distribution which can reduce material fracture.[63,64] The impact strength of PMMA reinforced with TiO2 (both on a micro and nanoscale) with concentrations of 0.1, 0.4, 0.7, 1, 3, and 5 wt.% showed that the incorporation of TiO2 had a significant effect on increasing the impact strength of the composite.[65] The impact strength profile is on the basis of a pure PMMA impact strength of 4.7 kJ/m^2.

Kumar et al. have investigated the impact strength of PMMA material reinforced with 1 wt.% TiO2 nanoparticle using conventional microwave and water bath techniques.[66] The test samples in groups I and II were PMMA without and with reinforcement, respectively, which were processed with a conventional water bath. Groups III and IV were the test samples for pure PMMA and PMMA with reinforced, respectively, which were processed by the microwave technique. They reported that the group IV test sample had the highest impact strength, which was followed by the group II sample. With the addition of 1 wt.% TiO2 nanoparticle for samples treated with microwave, the impact strength increased 6%, while with traditional water bath techniques, the impact resistance was increased by 16 percent by nanoparticle reinforcement.[66] They explained that the determination of the reinforcement concentration (1 wt.%) was due to a study conducted by other researchers who recommended strengthening the mechanical properties of composites by incorporating 1 wt.% TiO2 nanoparticle.[38,67]

Impact strength is greatly affected by the use of processing techniques. The water bath technique is an easy and inexpensive procedure. Also, this technique is reported to have frequent cavity formation resulting in low-impact strength.[68] The microwave technique was reported to be shorter than the water bath technique in the formation of materials.[69]

Particle effect on impact strength

The majority of biomaterials from PMMA-TiO2 composites are applied under impact loading conditions, so the most prominent test to evaluate the mechanical properties of dentures is the fracture toughness test. The fracture toughness of the denture was assessed based on the shape and strength characteristics of the impact test.[70] Several studies reported that TiO2 nanotubes can be effectively used to increase the fracture toughness of PMMA-based denture materials.

TiO2 nanotubes have been added to commercial PMMA and it is claimed to increase the fracture toughness of composites by the addition of 1wt.% TiO2 nanotubes.[46] They also attributed this increase in mechanical properties to an increase in the bond strength at the interface between TiO2 nanotubes-PMMA which causes external force transfer to occur effectively at the interface of PMMA-TiO2 nanotubes. In a different study, the mechanical properties of flowable dental composites reinforced with n-TiO2 were reported to have increased fracture toughness compared to those without n-TiO2 reinforced.[47] It was also reported that composites with 3wt.% n-TiO2 showed the greatest fracture toughness with the lowest flowability effects. A study with n-TiO2 concentration greater than 3 wt.% was conducted by Naji et al.[44] and found that PMMA was reinforced by 5 wt.% n-TiO2 through an alkaline hydrothermal process had significantly higher fracture toughness than that without reinforcement (control group). At 2.5 wt.% n-TiO2 strengthen, the fracture toughness was not remarkably superior to that of the control group and was not significantly below 5 wt.% n-TiO2.[44]

Microstructural Characteristics

At the 3 wt.% TiO2 concentration, it was reported that the nanoparticles were uniformly distributed in the host matrix, as reported by Alrahlah and colleagues.[45] However, it was also reported that some TiO2 NPs exhibited agglomeration at the nanoscale which is thought to be due to the tendency of the nanoparticles to reduce their contact surface with PMMA. Alhotan et al. have observed the fracture surface of 7 wt.% TiO2/PMMA specimens after flexural testing using a scanning electron microscope (SEM). It was reported that the fractured surface of the pure PMMA specimen exhibits ductile-type failure behavior with irregular areas and small nanopores. The reinforced nanocomposite showed the presence of particle clusters with small cavities which indicated that the nanoparticles were not uniformly dispersed.[23]

The morphology in the cross-section observed by Shirkavand and Moslehifard shows that the TiO2 with a concentration of 1wt.% has a higher distribution compared to the two other groups as depicted in SEM images.[31] They explained that increasing the content of TiO2 nanoparticles in the acrylic matrix from 1 wt.% to 2 wt.% caused small cracks. This phenomenon also confirms a previous study conducted by Nawaz and Rharbi in which cracks were observed without the use of stress.[71]

Dafar and colleagues evaluated the composite fracture surface on TiO2 (n-TiO2) nanotubes, after testing for fracture toughness. It was reported that the unreinforced composite (control), contained spherical nanoparticles. It was also shown that the reinforcing particles were distributed in the composite matrix in the n-TiO2 reinforced composite. However, composites with n-TiO2 functionalized silanes showed that the nanotubes in the composite matrix are tightly embedded (50).

Conclusions

The mechanical properties of PMMA filled with TiO2 particles are reviewed extensively in this article. In recent years a number of studies were performed with an emphasis on further improvements in PMMA's mechanical properties by alteration using TiO2 particles. Acrylic mechanical properties which include tensile properties, compressive strength, creep strength, hardness, impact strength, and fracture toughness, can be improved by adding TiO2 nanoparticles to PMMA.

The effective increase in TiO2 concentration is about 2.5 wt% and the mechanical performance of TiO2-PMMA decreases at the content of TiO2 nanoparticles beyond 3 wt%. More significant improvement of PMMA properties can be obtained by combining nanotubes compared to TiO2 nanoparticles. This is mainly due to a higher surface-to-volume ratio of nanotubes than the nanoparticles. Considering this review, further studies are suggested to further suppress the incorporation of TiO2 nanotubes as reinforcement. This study is very prospective in order to strengthen the mechanical strength of denture materials.

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Declaration of Interest

The authors report no conflict of interest.

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