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Mechanical Performance of Reinforced PMMA-Based Composites for Denture Materials: a Review

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Abstract – *The main acrylic used to manufacture dentures is Polymethylmethacrylate (PMMA). Its mechanical properties have been improved through the incorporation of various particles of both micro and nano size and fibers. This article aims to review mechanical properties of PMMA-based denture materials. Particular attention is paid to the effect of adding fiber, filler, and nanofiller on the mechanical characteristics of PMMA. This review is based on scientific reviews, papers, and abstracts as well as studies published between 1980 and 2020 concerning the effects of surface modification, reinforcing agents, fillers, and fibers on mechanical properties of PMMA as a denture material. Numerous studies show that the addition of fibers and fillers and surface modification can strengthen the mechanical performance of PMMA-based denture materials. The results also show that most studies are limited to in vitro research without bioactivity and clinical consequences.*
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***Keywords**: Mechanical Performance, Denture, Reinforcement, PMMA, Surface Modification*

Nomenclature

PMMA	Poly (methyl methacrylate)
MPTS	Methacryloxy Propyl Thoxy Silane
ZrO_2	Zirconium dioxide
MPS	γ-Methacryloxy Propyltrimethoxy Silane
PE	Polyethylene
PP	Polypropylene
OPEFB	Oil Palm Empty Fruit Bunches
Al_2O_3	Aluminum oxide
ND	Nanodiamonds
NPs	Nanoparticles
HA	Hydroxyapatite
TiO_2	Titanium dioxide

I. Introduction

The development of denture materials has been carried out since the early first century by using materials such as ivory, bone, vulcanite, wood, as well as vulcanized rubber. The introduction of Poly (methyl methacrylate) as a dentures materials began around the middle of the twentieth century. Since then, PMMA has been superior to all other denture base materials, and almost all dentures are made with acrylic base material [1]. Although several new light-activated materials, e.g. polystyrene and diacrylate urethane, PMMA proved to be an excellent material for full and partial prosthesis dentures. Various kinds of stresses including compressive, tensile, and shear, could really lead to early failure of denture base materials in practical uses [2], [3].

Intraorally, recurring chewing over time may lead to fatigue damage to the prosthesis base. Extraoral, dentures can also have high impacts when accidentally dropped.

Extra-oral impact fractures are caused by inadvertent tooth damage [4], [5]. The incidence of denture fractures is relatively high: within three years of manufacturing, 68% of dentures fail and the incidence is greater in partial prostheses than a complete ones [6]. Dentures are needed, especially for patients who have lost or damaged teeth. The biocompatibility, non-toxicity, suitability of tooth color, and mechanical properties have been investigated in many studies devoted to the development of denture materials. The two main materials that are widely developed as denture materials are metal, namely titanium, while polymethyl methacrylate (PMMA) is developed from polymer. Metallic materials are heavy, heat conductive, difficult to manufacture, and relatively expensive. Therefore, the metal denture material is not much developed. PMMA has been developed as a denture material because it is biocompatible, low toxicity, resembles the natural color of teeth, and has adequate mechanical properties. Even so, due to mechanical and physical properties, PMMA is not an ideal material. For example, PMMA contains moisture that, during use, is susceptible to a cyclic load failure and can compromise its mechanical characteristics [6]. In fact, many studies have reported that high levels of shrinkage, fragility and poor mechanical characteristics of PMMA restrict its development to broad clinical applications [7], [8]. Many studies are currently focusing on the mechanical strengthening of PMMA to overcome its disadvantages and enhance its mechanical characteristics [3], [9]-[11]. It is crucial to understand that property in order to truly comprehend and interpret these recent changes. This review examines various reinforcements of dentures based on PMMA, including surface modification by means of a silane coupling agent as well as plasma, incorporation of

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fibers and particles in PMMA, and the influence on reinforcement on mechanical properties.

The rest paper is divided into five sections. Section I describes the activation of the polymerization mechanism for various types of PMMA including cold-cured, heat-cured, light-cured, and microwave-polymerized. In Section II, the mechanical properties of PMMA are explored related to its application in biomedical applications, especially for dentures. Section III explains the surface modification of PMMA using a silane coupling agent and plasma. In Section IV, mechanical reinforcement of PMMA by fibers is described by incorporating glass, polyethylene, polypropylene, polyamide, and natural fibers in host PMMA. Section V, the mechanical reinforcement of PMMA by particles, including alumina, silver, nanodiamond, silica-based particles, and hydroxyapatite.

II. Activation of the Polymerization Mechanism

The suspension or emulsion polymerization of PMMA is routinely produced [12]. The dental PMMA is formed by polymerization of the suspension. The addition of PMMA with the initiator, usually benzyl peroxide, consists of polymerization of the multimethacrylate monomers [13], [14]. Benzoyl peroxide is transformed into free radicals after being damaged by chemicals or heat generated.

These free radicals act on the methacrylate vinyl group and open up a dual bond to form a new single Carbon bond [15], [13]. Denture base acrylic resin is used in dental practice for many kinds of intentions. That resin can be classified based on the initial factors that trigger the polymerization reaction, namely heat, chemical polymerization materials, light, and microwaves [16], [15]. These factors greatly affect the properties of the material being produced.

II.1. Cold-Cured PMMA

The polymerization reaction in PMMA does not require heat, the reaction process occurs automatically which starts when the powder and liquid components are mixed. Most of these curing methods produce low molecular weight, leading to a rise in the number of unsustainable resin monomers, poor strength, and a drop in the temperature of the glass transition [17], [18]. In fact, this curing method produces little internal stack strain because no external heat source is used, however, the material is very sensitive to creep which can lead to denture distortion when used [19].

II.2. Heat-Cured PMMA

Heat-cured PMMA was a widely used material for denture based [20], [21] due to its superior properties in mechanical and chemical. The PMMA part is supplied in two components namely powder and liquid, which, on mixing and subsequent heating from a rigid solid. The powder portion is made up of PMMA, benzoyl peroxide as act an initiator, dibutyl phthalate as a plasticizer (to make it softer and add flexibility), titanium and zinc oxides as opacifiers, and pigments. The other portion is a liquid, such as the monomer of MMA, the cross-linking agent (ethylene glycol dimethacrylate), and hydroquinone [22]. A cross-linking agent is used to improve material characteristics.

The chain of MMA polymers is interconnected at different points and forms connections with neighboring chains due to two sites.

II.3. Light-Cured PMMA

Similar to resin-based restorative composites, Light-cured PMMA functions when subjected to visible light [23], [24]. The photo-sensitive agent will be activated to replace the traditional precursor with a free radical agent when exposed to light. The objects should be exposed to visible light for both the necessary period after adaptation to cast and teeth positioning in order to fully cure PMMA.

The light-cured PMMA can indeed be completed like a traditional heat-cured PMMA after polymerization [25], [26]. The light-cured PMMA provides the prospect of simpler manufacturing and fully controlled curing to allow enough time for handling and adjustment until the polymerization begins [27]. In comparison to the heat- and cooled-cured PMMA, the presence of residual monomers is lower than the possible benefits of light-cured PMMA.

Nevertheless, light-cured PMMAs, such as their limited cure depth, and technical sensitivity, are not widely used due to their inconvenience [28]. Light-cured PMMA has mechanical characteristics that are significantly less than traditional PMMA, so their uses are restricted to denture base repair [22].

II.4. Microwave-Polymerized Polymers

In 1968 the polymerized microwave was initiated and had become a favorite option for the polymerization technique for hot water baths [29]-[31]. The many benefits have been noted for that technique; short reaction times, a more clean procedure, and better casting adjustment [32], [33]. In addition, the number of retained monomers was found to be decreased for the microwave-polymerized resin [34]. In microwave polymerization, the increase in heat occurs due to the impact between molecules that are accelerated in high-frequency electromagnetic fields.

Since the heat generation phenomenon occurs in the material, no transfer into the material is required [35], [36]. Nevertheless, the microwave polymerization technique has some limitations. Excess heat, as well as fast heating, must be averted because the monomer may volatilize and trigger porosity, as already confirmed by regular PMMA resins whenever the polymerization of the microwave has been used [29], [37]. Different PMMAs made for this technique are accessible on the market.

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III. Mechanical Properties of PMMA

Depending on the biomedical use, the materials on PMMA should have some desirable characteristics. PMMA materials have therefore been updated and investigated widely with regard to many biological [38], [39], and mechanical [40]-[42], [10] characteristics. It is fundamentally essential for the denture base implementation to understand the best possible properties of PMMA. Since prosthodontic restoration must be conducted in a complex oral setting, in order to prevent inflammation of oral tissue and toxicity, PMMA in basic prosthesis materials must be biocompatible [43]. PMMA must be chemically extremely insoluble in saliva and oral fluids. It will, however, chemically bind the teeth so that they are not nutrient-sensitive. PMMA must be mechanically well-suited for chewing forces (e.g. stiffness, impact strength, and fatigue strength) [44].

The basic teeth materials undergo complicated chewing pressure in the oral cavity. In order to enhance the dentures' physical ability, superior mechanical properties are required for the material. Since the mechanical characteristics of materials can be substantially modified, various studies have been carried out to change the mechanical characteristics of PMMA, including impact strength [45]-[47], flexural strength [3], [10], [48], and fracture resistance [49], [50]. Denture materials need fracturing resistance which is the material's ability to resist spreading cracks through notches or defects on the surface [51]. The fracture toughness of heat-cured PMMA was 26.4% greater than that of cold-cured PMMA (about 1.53 $MN/m^{3/2}$) [52].

Therefore, heat-cured PMMA was more effective at inhibiting crack propagation. Surface defects due to wear can act as a notch that triggers a stress concentration that causes the crack to propagate effectively which ultimately reduces the impact strength [53]. Many studies have reported that butadiene-styrene can significantly increase the impact strength of PMMA [54], [55].

However, the wear resistance of PMMA is lower than that of alloy castings and dental porcelains [56], [57].

IV. Surface Modification of PMMA

IV.1. Surface Modification with Silane Coupling Agent

The interface plays a critical role in dispersing the surface layer, matrix and filler, and dependence when going to work in multiple component systems.

Consequently, the surface properties must be adapted to increase adhesion. Coupling agents are conformity boosters for the chemical unification of dental equivalent products. In adhesion enhancement between resin composites and other indirect restorative components, silanes are very successful. Polymers have been strengthened by inorganic filler particles. Many of these particles are hydrophilic and aglomerating in a hydrophobic setting similar to PMMA [58], so coupling agents are utilized in order to enhance the compatibility of fillers with organic matrices [59].

Several works have developed different methods of silane treatment as particle surfaces modifiers and the most common saline coupler is three-methacryloxypropyltrimethoxysilane (MPTS) [60], [61], [41]. Hydrophilic and hydrophobic ends are bipolar molecule MPTS. It may bind with its hydrophilic end to the inorganic filler particle. The hydropathic end of MPTS consists of alcohol-free copolymerized groups that bind with the neighboring MPTS molecules [62]. A relationship between a variety of surface modifications on the mechanical properties of silicone denture liner auto polymerization on dentures was studied by Atsü and Keskin [63]. Two PMMA blocks have been put in molds and the materials for soft lining have been packed into space and polymerized. Before the tensile examination, both specimen groups were thermocycled (5,000 cycles).

Tensile test data has been obtained by one-way ANOVA test and Duncan's test. The finding indicated, compared to the control group, that silicone-based soft liner did not enhance its resistance to surface processing compared to the silane-modified surface. However, a different study reported that the nanometer ZrO_2 was effectively modified with the silane coupling agent Z-6030 at a dose of 3,5 percent and that the flexural strength of PMMA/ZrO_2 nanocomposites was markedly improved [64]. To obtain an alternative coupling agent for silane, a titanate coupling agent is used in producing PMMA-filled nano barium titanate composites [65]. The titanate coupling agent has successfully activated the nano barium titanate. A single layer is formed in the titanate coupling agent surface, which increases the nano barium titanate dispersion, promoting reduced porosity levels and increasing the composites' fracture toughness, indicating that the titanate coupling agent is better than silane.

IV.2. Surface Modification by Using Plasma

Plasma treatment may be one of the most flexible surface treatment methods used in many different fields such as materials for medical equipment for the improvement of adhesive properties [66]-[69]. The specific surface characteristics needed for various applications can be used with oxygen, nitrogen, and argon in a plasma chamber. For instance, oxygen-plasma treatment may improve surface energy, while fluorine-plasma treatment reduces surface energy and gives the chemistry of polymer facelift. Liebermann et al [70] reported a significant improvement of surface energy, however without effect on surface roughness, in the polishing and air-obscured plasma modified based on PMMA. The untreated plasma group had a greater strength of the tensile bond than the plasma group. The plasma-treated PMMA did not enhance adherence to the adhesive resin composite cement. Plasma surface treatment is a well-established procedure since the polymer surfaces cannot be modified without changing their bulk characteristics. The chemical functionality, surface state, weight resistance, and binding capacity of polymer surfaces have been shown to be significantly

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altered by plasma processing [71], [72]. To date, there were few works that investigated the impact of plasma therapy on dental resin adhesion [73]-[75]. With regard to plasma therapy for silicone rubbers/PMMA, Zhang et al [76] report that a tensile bond strength has been successful in enhancing by 2 minutes of oxygen plasma therapy for the base acrylic denture liner, particularly in the time that specimens were uncovered to air for 12 hrs thereafter plasma therapy. In a different work, the surface characteristic of PMMA prior to plasma treatment for various time spans was examined by Zhang et al. [77] and many transformations in the chemical structure were reported. Bicer et al. [78] indicated that the bond strength values of argon plasma therapy with different exposure times between the silicone denture liner and self-polymerized acrylic resins have changed. In addition, another study indicates that, under oxygen plasma treatments with long exposure periods, adhesion between PMMA and denture liner is enhanced rather than argon plasma [75].

V. Mechanical Reinforcement of PMMA by Fibers

Dentures made of acrylic resin are subject to fractures due to impact loads and it has even been reported that 68% of the incidents occurred several years after fabrication. In response to this problem, metal wires have been selected as reinforcement in the base matrix PMMA denture. However, the produced composites had unexpected mechanical performance due to poor bond strength at the PMMA matrix-reinforcing interface. The impact resistance of the PMMA matrix composites has been successfully increased by incorporating the rubber phase into the matrix although this modification increases production costs. Dentures made of acrylic resin are subject to fractures due to impact loads [79], [80] and it has even been reported that 68% of the incidents occurred several years after fabrication [81]. In response to this problem, metal wires have been selected as reinforcement in the base matrix PMMA denture [82], [83]. However, the produced composites had unexpected mechanical performance due to poor bond strength at the PMMA matrix-reinforcing interface. The impact strength of the PMMA matrix composites has been successfully increased by incorporating the rubber phase into the matrix although this modification increases production costs [84]. The mechanical performance of the PMMA material as a denture base is improved by adding non-metallic fibers due to its biocompatibility, aesthetic value, and mechanical properties [85]. The fibers that are widely used include glass fiber [86]-[88], polypropylene fibers [89], aramid fiber [90], [91], carbon fibers [13], [79], nylon fibers [92], [93], and polyethylene fiber [94], [46].

V.1. Glass Fibers

Glass fibers have been widely used as reinforcement in biomedical materials. Its application as reinforcement to PMMA-based imitations demonstrates superior mechanical performance in tensile, impact strength, and material hardness [45], [86]-[88]. It was also reported that the presence of glass fibers in PMMA-based dentures reduced deformation by less than 1% [86]. Agha et al. [95] have reported that dentures are highly influenced by the adjustment of the orientation of the fibers in the host matrix. Thus, placing the glass fibers on the side near the composite surface that is subjected to tensile stress, can strengthen the flexural properties of composites. The glass fibers positioning, however, only increases flexural toughness and flexural modulus on the non-neutral side of stress and compression, respectively.

Unidirectional glass fiber-treated curing γ-methacryloxypropyltrimethoxysilane (MPS) solutions have been added within auto polymerizing PMMA. After approximately 120 minutes of cure, the PMMA-glass fiber composite was of the highest transverse force (approximately 152 MPa). However, MPS polymerization above 150 °C may lead to the lowest composite strength, which is approximately 91 MPa.

Scanning electron microscope observations on the fiber-matrix interface show that good bonding occurs in PMMA which is treated at a temperature of 100-150 °C [96]. Furthermore, Vallittu [96] concluded that the reduction in transverse strength of glass fiber-PMMA composite is due to improper impregnation of PMMA, not by bad surface bonding.

V.2. Polyethylene and Polypropylene Fibers

Polyethylene (PE) fibers-reinforced PMMA that was either surface treated or not could significantly increase the impact strength [97]. Furthermore, an increase in the modulus of elasticity and toughness of PMMA can be obtained by strengthening the woven PE fibers.

However, in previous studies, it was reported that the preparation of woven PE into the PMMA matrix could not be done easily [98]. Polypropylene (PP) fibers were shown to significantly improve the impact strength of PMMA, especially if the PP fiber surface was treated with plasma to strengthen the PP fiber-matrix bonds and reduce the occurrence of fracturing [89]. Mathew et al. [88] have conducted a study on strengthening PMMA by PP fibers with weight variations of 2.5, 5, and 10 wt.%. It was reported that at a weight concentration of 10% PP fiber with a length of 12 mm, the impact toughness of PMMA could be significantly improved. A recent study by Ismaeel et al. [99] shows that the incorporation of salinized PP fibers in heat-cured PMMA improves impact strength, and tensile, but decreases wear resistance dramatically.

V.3. Polyamide Fiber

Nylon is a polyamide fiber that is commonly found as a reinforcement for denture base resins [100]. The presence of this nylon fiber increases the flexural properties of both the flexural modulus and the flexural

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strength [101]. However, increasing the concentration of nylon fibers was reported to decrease the hardness but increase the fracture resistance of the denture resin [102], [103].

However, modification of the polyamide is needed to improve its properties. Unfortunately, little literature can be found on the clinical performance of polyamides, therefore, it is highly recommended that a more in-depth and careful study be undertaken in particular in patients rejuvenated by polyamide prostheses [93].

V.4. Natural Fibers

Many natural fibers have been explored in order to strengthen the dentures, such as Oil Palm Empty Fruit Bunches (OPEFB), hemp fibers, and bamboo fibers [104], [105]. The application of OPEFB has revealed that the flexural properties (modulus and strength) of acrylic resin have become greater considerably [104]. However, it was found that a weak interface bond could reduce flexural strength.

Oleiwi and co-workers [105] carried out studies into the mechanical characteristics and the influence of siwak and bamboo fibers on PMMA by varying its fiber weight fraction from 3% up to 9% and fiber length up to 12 mm. PMMA has been supplemented with alkali-treated fiber and manually laying the composite.

The impact strength by weight fraction of the fibers was indicated to be decreased, but the pressure strength was increased to 530 MPa by 9 wt.% fibers. But the impact strength can be enhanced by lengthening the fiber despite this further increase as the reason for the decrease in the compressive strength of the composites.

VI. Mechanical Reinforcement PMMA by Particles

The main concern of orthodontists and health care providers' usage of detachable acrylic devices is the deposition of plaques as a result of structures and surface morphology going to lead to the microbial load of cariogenic bacteria [106].

Several methods were developed to focus on improving resin equipment's mechanical performance and reducing the probability that caries form. Self-cleaning antimicrobial compounds have been widely used in order to improve the propensities of a material and its antibacterial properties by using various nanofillers as the orthodontic agent [107]-[110].

A variety of nanoparticles have been applied to various biomedical materials for the purposes of inducing and mechanically enhancing antimicrobial action. Various fillers both derived from ceramic and metal particles have been examined for PMMA material enhancement purposes [111]. The ZrO_2 nanotube, for instance, showed more impressive mechanical strength than the ZrO_2 particles.

Likewise, the addition of metal nanoparticles improves denture base heat capacity [112], [113].

VI.1. Alumina Reinforced PMMA

Biocompatibility was achieved by incorporation of Al_2O_3 powder in the heat-cured PMMA. Due to the silane-processed aluminum particles, the mechanical properties have improved considerably, mainly the strength of compression, flexurization, and wear [114], [115]. The PMMA surface roughness is not significantly affected.

PMMA has significantly improved thermal conductivity [116], [117]. The improvement of Al_2O_3 is limited primarily by the coloring of the resin [118], [119].

VI.2. Silver Reinforced PMMA

The compression of PMMA is enhanced due to its various metallic nature by the incorporation of silver particles [91]. PMMA was complemented by silver and graphene in nanosize particles to substantially rising tensile, compressive, and flexural strength [91].

However, Kul and co-workers [120] confirmed that due to the introduction of silver particles, the flexural strength does not change considerably.

VI.3. Nanodiamond Reinforced PMMA

The application of nanocomposite polymers may provide the bulk polymer with high strength [121]. The nanodiamonds (ND) are biocompatible and have gained momentum since these nanoparticles have outstanding properties including high strength, corrosion resistance, and good thermal conductivity [122], [123]. The ND particles are bioactive and many studies incorporated them as reinforcement on PMMA [124]-[127]. Protopapa and colleagues [125] demonstrated having improved the impact strength and the modulus of Young around 2.084 GPa by integrating about 0.83 wt.% ND nanoclusters into PMMA. Mangal and colleagues [128] have assessed the effect of incorporating 0.1, 0.3, and 0.5% ND into PMMA in terms of flexural strength and modulus of elasticity compared to control nanoparticles and zirconium oxide.

The hardness of the surface is also assessed. The results showed that the mean bending strength and the modulus of elasticity were statistically significantly higher in all ND-PMMA nanocomposite groups than in the other sample ones. In comparison to the control group, the Vickers hardness value increased significantly higher at 0.3 wt.% and 0.5 wt.% ND.

VI.4. Silica Based Particles Reinforced PMMA

Nanosilica is one of the most widely used kinds of nanoparticles (NPs) in science. Some works have examined nanosilica particulate matter in addition to PMMA resins. Silica NPs were successfully introduced within PMMA. Some works have reported significant impacts on the mechanical and thermal characteristics of such materials [129]-[131]. Nevertheless, inappropriate types or dosages of loading of NPs can lead to reduced mechanical characteristics [132], [133]. The incorporation of nanosilica into PMMA has been investigated in several

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studies [133]-[135] and it has been reported that the flexural strength, hardness of the material, and crack resistance are affected by the presence of the nanosilica.

They also reported that at the nanoparticle weight fraction below 2%, the particle dispersion was the best and effectively improved the properties of the composite. At nanoparticle content above this limit, agglomeration and poor dispersion occurred entire part of the material.

Moreover, the limited Van der Waals forces in the aggregates make it easy for aggregates to break easily when subjected to lower loads which cause the crack propagation to be unstable [136]. PMMA/nanosilica were tested in Yang and Nelson's [137] work on stresses and nanoparticles were added to increase the tensile strength by over 50% and almost double the modulus of elasticity, while keeping going on elongations at rupture. Natural silica NPs were already chosen to increase PMMA's properties, as a biologically compact material that has a high resistance to fractures [132]. Recently the incorporation of nano-inorganic materials has gained wide attention to improve the performance of the polymer.

VI.5. Hydroxyapatite Reinforced PMMA

Numerous work has recently been done to examine the effect of introducing hydroxyapatite (HA) in PMMA as a bone substitute material. Although HA has been widely used as a biomedical material including bone and dental implants, there are still few studies investigating the principle of HA for reinforcing PMMA-based denture materials. However, the introduction of HA particles into the PMMA matrix has been reported to convert initially non-bioactive PMMA into bioactive composites, and at the same time improve mechanical performance, particularly surface hardness and modulus of elasticity [138]. In a three-point bending test, compressive compression, and wear test on nano HA reinforced PMMA composites, Zebarjad et al [139] reported that adding nano HA did not find a directly proportional relationship between the bending test results and the level of HA content. Increasing the HA fraction up to 10 wt.% was reported not to significantly change the flexural properties of the composites. An investigation conducted by Shyang et al [140] showed that the presence of HA NP reduced the strain and flexural strength of HA/PMMA nanocomposites. However, in other studies, it has been reported that the incorporation of HA NP can increase the E-modulus and flexural strength of nanocomposites [141], [142]. Even though the HA particles can to some degree be added to PMMA to enhance the physical characteristics of composites, the filler-matrix interface bonding needs to increase for strengthened the mechanical performance such as strain to break and crack propagation resistance. The addition of nanoparticles may lead to the formation of agglomerations that reduce the surface area of the particles covered by a matrix, which will weaken the mechanical strength of the material [143], [144]. The powder samples for 5, 10, and 15 wt.% of HA/PMMA are shown in Figs. 1 [142].

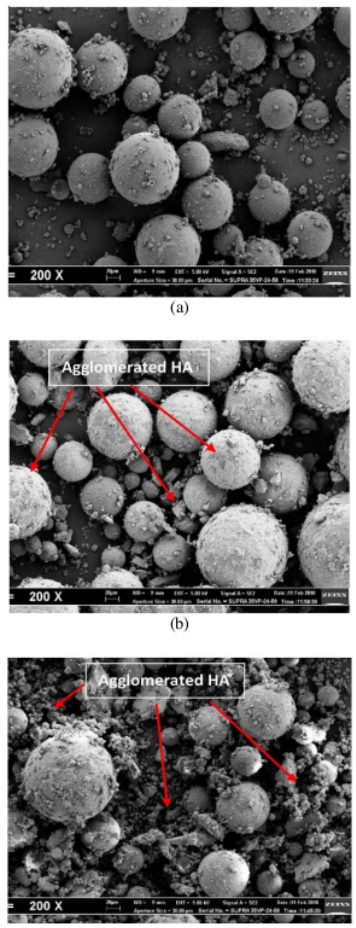
VI.6. ZrO_2 Reinforced PMMA

Some studies are designed to improve the mechanical characteristics of PMMA, including the use of bioceramic nanoparticles as reinforcement due to their unique properties [145]-[147].

Zirconia (ZrO_2) is one promising material commonly used in numerous restorative dentistry, including crowns, and orthodontal braces [148].

Zirconia has a strong mechanical characteristic with flexural strength, hardness, and fracture toughness is approximately 1000 MPa, 1200 HV, and 10 MPa $m^{1/2}$, respectively [149]-[151]. In contrast with other ceramic materials like alumina, zirconia demonstrates great biocompatibility [145], [150], [152].

A few works indicate a substantial improvement in its flexural strength through the introduction of Zirconia (ZrO_2) into the PMMA matrix [112], [120], [153]-[155].



(a) (b) (c)

Figs. 1. SEM images taken on powder samples for 5 wt.% HA/PMMA (a), 10 wt.% HA/PMMA (b), and 15 wt.% HA/PMMA (c) [142]

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Nevertheless, there was a slight reduction in flexural strength; this could come from the particle grouping in the host matrix.

Moreover, the fracture properties, as well as the PMMA hardness [112], [156], have been substantially improved by the incorporation of ZrO_2. Also, neither the impact strength nor the surface hardness of the PMMA reinforced by ZrO_2 did not increase significantly compared to the non-reinforced ones. There was also a study that reported that the impact strength and surface hardness are decreased by the presence of zirconia [155]. The hardness performance in various weight fractions of ZrO_2 was shown in Fig. 2. Fracture tightness is one important parameter as it marks the initiation of cracks and prevents the material from spreading cracks [157].

Various factors can cause a decrease in the fracture toughness of PMMA/ZrO_2 nanocomposites by increasing the concentration of fillers, particle agglomerates, particle dimensions, and the chemical response of polymer surfaces to particles [100], [112], [158]. In the case of hardness, some studies [157], [159] show that PMMA/ZrO_2 hardness was increased by increasing ZrO_2 content up to 15 wt.%. The incorporation of hard-yttrium stabilized nanoparticles could lead to increased nanocomposite hardness. The higher hardness will increase the abrasive wear resistance of composites.

However, following water immersion, surface hardness decreased leading to the removal of residual monomers and at the same time water absorption [159]. The deflection characteristics in relation to the bending force under cyclic loads at 3 wt.% and 5 wt.% zirconia contents as compared to the control composites are shown in Fig. 3.

VI.7. Mechanical Reinforcement PMMA by Titanium Based Material

In the manufacture of implants for dental applications, Titanium (Ti) and its alloys are choice materials. Because of their low hardness, these biomaterials provide comparatively poor tribological characteristics.

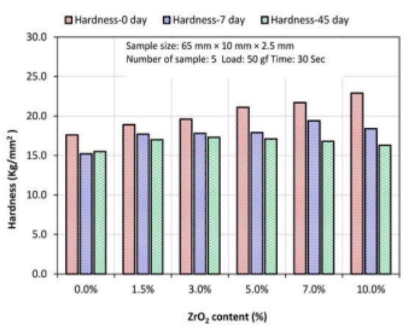

Fig. 2. Vickers hardness performance in various weight fractions of ZrO_2 for PMMA/ZrO_2 nanocomposite after dyeing for 0, 168, and 3480 hrs [155]

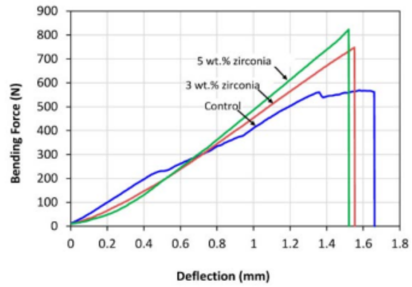

Fig. 3. A characteristic curve of bending load - deflection in the absence of fatigue under cyclic loading for neat PMMA and PMMA reinforced by zirconia nanoparticles [3]

Modifying the chemical composition, it can change the biological characteristics of Ti alloys. Another method that can be used for this purpose is to manufacture Ti composites in which the other constituent components are biocompatible and have excellent mechanical properties [160], [161]. Ti fillers will theoretically increase the mechanical strength of the composite [162], [163] by adding them as an additional component. Titanium dioxide (Titania, TiO_2) is often recommended in orthodontics due to its color similar to a natural tooth, and its excellent biocompatibility [164], [165]. Also, the mechanical properties of TiO_2 nano-particles are excellent. For instance, they have an elastic modulus of about 230 GPa. Titania has also been an adequate dental antimicrobial additive, with other qualities including white-colored powders, nontoxic, and chemical stability, and also low costs [165]. Since titanium remains stable in the body and degradation resistance, bioactive titanium can thus remain stable within the body. It was then developed a composite bone cement containing n-TiO_2 which has high mechanical performance. Among many particles incorporated into the PMMA denture base, TiO_2 nanoparticles (NPs) have been extensively investigated due to their remarkable durability, catalytic effect, accessibility, color white, efficiency, and low cost [166], [167]. TiO_2 NPs are also highly resistant to corrosion, chemical stability, and non-toxic [168]. The profile for PMMA and 2 wt.% TiO2/PMMA observed by differential scanning calorimetry and thermogravimetric analysis was shown in Figs. 4 [110]. In order to improve its mechanical and biological characteristics, multiple studies have been carried out to evaluate the mechanical characteristics of PMMA in combination with TiO_2 NP [169], [170]. Based on those studies, the main fracture mode was considered to be the evaluation of the bending strength among those involved in the loading attributes of the oral dental based.

Many studies indicate that flexural strength increases at TiO_2 content below 5 wt.% [169], [171]. It was also reported that silanization of TiO_2 NP could increase the surface energy and better the interaction between the matrix and nanoparticles. At a concentration of 1%, the nanoparticles were dispersed and no particle agglomeration was found [172].

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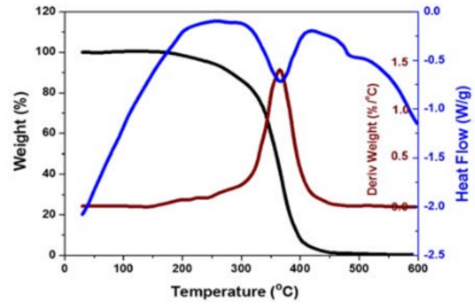
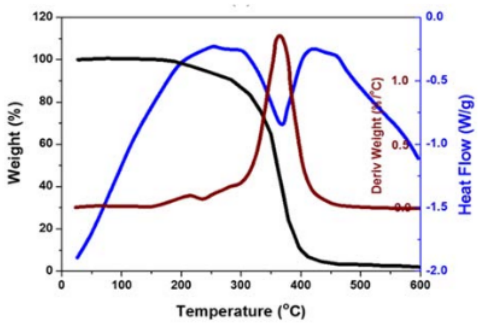


(a)



(b)

Figs. 4. The profile for (a) PMMA, and (b) 2 wt.% TiO_2 / PMMA on differential scanning calorimetry and thermogravimetric analysis [110]

The variations in flexural strength obtained with various TiO_2 NP concentrations can be attributed. Because nanoparticles have a very high volume-to-surface area, only a small amount of them are required to change the properties of polymers [173], [174]. Once the TiO_2 NPs join the matrix, the mobility of the polymer chains is reduced because the filler and matrix have strong interface bonding. This is possibly due to the reduction in polymer segment motion, the inherent TiO_2 NPs modulus, and a good filler-matrix interface bonding.

This phenomenon results in an increase in resin strength [175]. However, TiO_2 can act as impurities and interrupt polymer chains if their concentration exceeds 5 wt.%. The increase in filler concentration further reduces the polymer chains' movability going to lead to brittleness and premature fracture [176], [177]. TiO_2 NP may act as a highly concentrated plasticizer which can reduce the transformation rate of monomers thereby reducing flexural strength [177], [178]. In terms of tensile strength, the incorporation of TiO_2 NPs increases the tensile strength of nanocomposites by increasing the TiO2 NPs content up to 1 wt.% and then decreasing with increasing TiO_2 content added to the host matrix [177]-[180].

The TiO_2 NPs concentration above that value plays a role as a composite impurity that can reduce tensile strength. Mosalman et al. [181] reported the mechanical properties of PMMA-TiO_2 nanocomposites that the flexural strength was increased by 3.75% by the addition of 0.5wt.% TiO_2 content.

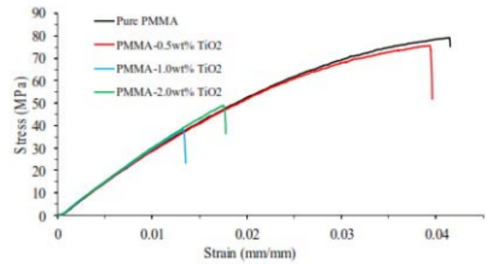


Fig. 5. Strain-Stress curve of tensile test for various TiO_2 content [181]

The impact strength and Young's modulus were also increased by the presence of 2 wt.% TiO_2 NPs in the host matrix, even the increase in impact strength could reach 229% compared to the pure PMMA value. Stress-strain characteristics for various Ti content are presented in Fig. 5. Shirkavand and Moslehifard [179] reported that 1 wt.% TiO_2-PMMA nanocomposite is a superior filling dispersion in cross-section. Small cracks in the matrix were found in the TiO_2 content increased from 1 wt.% to 2 wt.% where samples were prepared without applying pressure.

VII. Conclusion

In this article, the mechanical properties of PMMA-base denture composites were thoroughly reviewed. In dental clinics, PMMA products are frequently reported to decolorate, degrade, and fracture.

This indicates that PMMA properties need further enhancement. Much study has been undertaken in recent years with an emphasis on further improvement of the mechanical properties of PMMA. Surface modifications and the use of additional materials (fibers, nanofillers, nanodiamonds) involving chemical or mechanical reinforcement led to remarking advancements in mechanical properties such as impact strength, flexural strength, wear resistance, and fracture toughness.

However, the aesthetics, performance, and biocompatibility of denture materials in the oral cavity should also be a concern. Therefore, investigating the use of modified PMMA is very important to develop its application in dental clinics as well as in dental hospitals.

Additional studies on improved mechanical properties and greater aging resistance are required, so as to maintain an extended amount of time that the tooth material is prolonged. In addition, in vivo performance of the modified PMMA remains dubious and further study is necessary.

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Dwi Windu Kinanti Arti was born September 26, 1987 in Serang Regency, Banten province, Indonesia. She completed her master's degree in dentistry in 2015 at the Universitas Muhammadiyah Yogyakarta. Her research is focused on dental and oral health. Many books and research articles that have been published addressed dental and oral health. She and Dr. Purnomo are currently carrying out a project to develop denture materials and plasma applications in denture restorations. Dwi Windu Kinanti Arti, M.MR is currently serving as the Director of the Dental and Oral Hospital of Universitas Muhammadiyah Semarang. Also, she is a member of the professional organization of the Indonesian Dentists Association.

Ana Hidayati Mukaromah is an Associate Professor in Chemistry. She was born on May 02, 1965 in Semarang Regency, Central Java Province, Indonesia. He completed his doctoral study in 2017 in the field of Chemistry science from Institut Teknologi Bandung, West Java Province, Indonesia. His research is focused on ZSM-5 Membrane to Reduse Carbon Monooxide. Many papers in ZSM-5 Membrane were published in various international journals, including published in the Journal Of Mathemathical & Fundamental Sciences, and Proceeding Internasional Atlantis, in ALCHEMY Jurnal Penelitian Kimia. Currently, various researches in phytochemical test, antibacterial test, and antifungal test on hebal plants, test for Malondialdehid levels and identicication of Profil Protein were published in International Journal of Microbiology, Biodiversitas Journal of Biological Diversity, and Plasma medicine. Currently, she is working on a biomedical material development project, especially denture materials and test for Malondialdehid levels and histhopathology in Gastric White Rats exposed to Formalin and Treated with Grape Seed Exstract. Dr. Ana Hidayati Mukaromah has been a member of professional organizations including as a member of the Indonesian Chemistries Association (Himpunan Kimia Indonesia).

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