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Acrylic Composite Biomaterials for Dental Applications: A Review of Recent Progress

Mohammed Razzaq Mohammed[a*], Ahmed Namah Hadi[b]

[a]*Department of Mechanical Engineering, College of Engineering, University of Misan, Iraq*

[b]*Department of Biomedical Enginnering, College of Engineering, University of Babylon, Iraq*

[a]*Email: m.r.alassadi14@gmail.com*, [b]*Email: ahmed.hadi.eng@uobabylon.edu.iq*

Abstract

Polymethyl methacrylate (PMMA) is regarded as one of the most widely used and prominent biomaterials for biomedical applications, particularly in dental technology. The unique properties of PMMA regarding its aesthetics, price-affordable availability, easy manipulation, low density, and tailorable mechanical behaviours, make it an eminently suitable biomaterial for such applications. Despite its beneficial properties, PMMA has also some shortcomings in terms of susceptibility to hydrolytic degradation and having insufficient mechanical properties that could prevent this material to be able to handle the various applied forces during its use, which in turn make it vulnerable to fracture. Furthermore, PMMA could serve as a substrate to growth of harmful bacteria and fungi such as *Candida glabrata* that can cause agents of oral cavity infection and could seriously affect the stamina and person’s health in general. A wide range of approaches have been developed in order to enhance not only the mechanical and thermal behaviours but also water sorption, solubility and the biological activities of PMMA. Incorporating of reinforcement additives into PMMA matrix can improve these properties of PMMA. Several methods and materials have been utilised to reinforce acrylic resin denture base. One of these methods is the reinforcement by using particles whether from natural or synthetic sources including metals and ceramics. Apart from their sources, the particles surface characteristics, quantity and level of dispersion play an essential role in overall behaviour of the composites. Other types of reinforcements are natural and human-made fibers. Each of which has merits and disadvantages; while the synthetic fibers can provide better mechanical properties, natural fibers promote creating better composites in terms of biocompatibility and affordability. Nanotubes are an other spectacular kind of materials being utilised in some studies as a reinforcing phase for PMMA composites. This review will highlight the recent studies that have been conducted for the last decade regarding the development of PMMA-based composite biomaterials for dental applications.

Keywords: Polymethyl methacrylate (PMMA); Composite Biomaterials; Dental Applications.

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* Corresponding author.

1. Introduction

Polymethyl methacrylate (PMMA) is a lightweight, nondegradable synthetic polymer. It is utilised in abroad variety of biomedical applications that require permanent, mechanically stable structures to be commonly applied for non-metallic implant material in orthopaedics and bone tissue regeneration (1-4). It is used as a common bone grafting material, mainly in the fixation of orthopaedic prosthetic materials for hips, knees, ankle and shoulders. This polymer is also considered to be perfectly proper in spinal and oncologic surgery applications (5, 6), and for treatment of craniofacial tissue defects such as skin (1, 7). Apart from the high stability and long lasting properties of PMMA, its excellent biocompatibility and hemocompatibility, and optical properties make it a promising candidate for applications such as blood pumps, dialyzers and implantable ocular lenses and hard contact lenses (2, 8). PMMA has also been attracted considerable attention to be potentially used in prosthetic foot (9-11). Besides, PMMA resin has also a wide range of applications in dental technology such as the fabrication of artificial teeth, interim-fixed restorations, dentures, provisional crowns, and occlusal splints (12-15). Various types of PMMA are available namely, heat curing PMMA, cold curing PMMA and light-cured or visible light cured PMMA. Each of them has different biological and mechanical behaviours make it utilised in specific dental applications that could be different than other PMMA types. Heat curing PMMA materials can be available as powder and liquid and be widely used for construction of denture bases and dentures (16). While PMMA powder and liquid both contain initiator of different materials, the former also composes of plasticizer, opacifiers, fibers, and pigments or dyes. On the other hand, the PMMA liquid contains, additionally to the initiator, methyl methacrylate (MMA) monomer and a cross-linking agent (17). With this type of PMMA materials, heat energy that could be generated from a water bath is required to promote the starting of polymerisation reaction of the components. Heat-polymerised acrylic resin has excellent appearance, simple processing technique and easy repair; nevertheless, its insufficient mechanical properties such as impact strength and transverse strength make it not be able to handle the applied forces during clinical application, leading to the fracture and failure. Microwave energy is another source of heat energy that can be used to polymerise and cure PMMA (13). Microwave curing has a short curing time which could be in a few minutes compared to hours of heating and cooling requiring in the conventional heat curing cycle (18, 19). The physical properties of microwave PMMA materials are comparable to conventional heat-cured PMMA (13). Nevertheless, these materials cannot be polymerised in the same way of that for heat-cured PMMA because of the absent of the initiator that is typically present in microwave-cured PMMA (19). The weak bond strength of microwave-cured PMMA (13), and the expenses and potential fracture of the used equipment and non-metallic flasks are the main limitations accompanied with the usage of these materials (20).Cold-cured PMMA or auto-polymerising PMMA does not require thermal energy, and the initiator used with this type is typically a tertiary amine (21). Cold-cured PMMA has better dimensional stability and minor polymerisation shrinkage, but a remarkably lesser degree of polymerisation than heat-cured PMMA (16, 21). In addition to uncured residual monomers in the polymerised material and inferior mechanical properties, discoloration can be caused with cold-cured PMMA due to oxidization of the amine initiator with time (21). Due to these limitations as well as their lower glass transition temperature (22), cold-cured PMMA materials are currently only employed in a narrow range of dental related applications (16). The other kind of PMMA is light-cured or visible light cured PMMA, which is cured when exposed to visible light in the present of a photosensitive agent (16). The light-cured PMMA has many merits over other PMMA kinds in terms of easier fabrication, full control of curing (13) as well as reducing re-polymerisation shrinkage, residual monomers, and bacterial adhesion (23). However, restricted limited curing depth, relative difficult manipulation, and cost are some of the disadvantages of light-cured PMMA materials that make this type of PMMA not widely utilised (20). The prominent use of PMMA in dental industry is due to its high biocompatibility, processability, appropriate aesthetics appearance versatility in terms of shaping, and capability of being easily fabricated and machined (24-27). Nonetheless, PMMA used in some dental technologies neither displays proper antimicrobial properties (28) nor shows optimum mechanical behaviours such as the flexural, transverse and impact strength (26, 29-31). Even though PMMA has been used in dental applications for many years, efforts have been made to enhance its properties. These include thermal and mechanical properties, water sorption, appearance, and solubility. A striking approach of making such enhancements is by reinforcing PMMA with biomaterials for engineering on demand composites. PMMA composites made of incorporating either natural or synthesis particles, fibers, or nanotubes for potential use in dental applications will be highlighted in this review. Figure 1 illustrates some of the widely used reinforcing materials for PMMA denture base material.

Figure 1: Examples of materials utilised for reinforcing PMMA. From left to right: titanium dioxide nanoparticles, alumina nanoparticles, glass fibers, polyester fibers.

2. Particle-Reinforced PMMA Composites

A wide range of particle-reinforced PMMA composites have been developed to improve PMMA denture base material properties. As in the particle-reinforced composites used in many applications, a number of parameters can influence the performance of particle-reinforced PMMA composites. Particles type, geometry, content, dimension, and distribution play major roles in the resulted composite behaviour. Furthermore, the composite properties can be tuned by controlling the interaction between the particles and the matrix. Thus, particle surface modification seems to be considered when there is a lack of adhesion between the particles and matrix. A quite variety of particles have been available for reinforcing PMMA denture base material; these include: natural particles, noble metals and ceramic fillers (Figure 2).

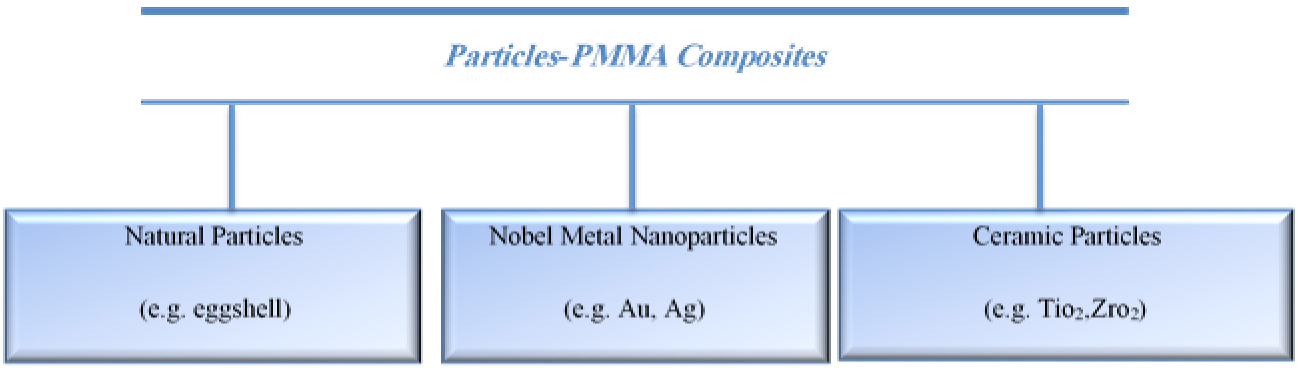

Figure 2: Types of particles used for making particulate reinforced PMMA Composites.

2.1 Natural Particles

Particles that are derivative from natural sources such as plants can contribute in reducing composite cost as well as reducing the harmful effect of using synthetic materials on the environment. Various natural particles either in micro or nano size have been added to PMMA to fabricate dental composites. The influence of adding pistachio shell powder into PMMA was investigated by Oleiwi and his colleagues who used hand lay-up process to fabricate a composite from these materials. The powder was added in different weight fractions and average particle sizes up to 12% and 212µm, respectively. It was found that the composite material with 9%wt particles had the highest compression strength and hardness among the other formed composites (32). Apart from improving the mechanical properties of denture acrylic resins, it is greatly preferred usage of materials that can be costly affordable. For this purposes, novel composite materials were formed from eggshell powder and PMMA resin using the hand lay-up method. The values of tensile properties and fracture toughness were enhanced after the incorporation of 7% weight fraction of eggshell powder (before and after burning). This ratio, on the other hand, resulted in a decrease in elongation percentage at break and impact strength (33). Hardness and tribological properties of PMMA based denture composite that was reinforced with various ratios of seashell nanopowder were evaluated in a different study. The addition of 2% seashell nanopowder had no substantial effect on the Vickers microhardness; however, increasing the filler nanopowder content up to 12wt% resulted in improving the microhardness and wear resistant of the PMMA/seashell composites (34). Because they contain collagen and hydroxyapatite (HA) minerals, fish scales were used to reinforce the acrylic resin. The flexural strength of heat cured acrylic resin (HcAr) was illustrated to decrease after addition 5% and 10% of fish scale powder (35). Two different natural nano-powders namely, pomegranate peels (PPP) and seed powder of dates Ajwa (SPDA) were utilised apart with various weight fractions to reinforce PMMA resin. A considerably improvement in the strength for both fabricated composites in comparison to bear PMMA. Moreover, PMMA/nano PPP composites had higher properties as compared to the PMMA/ nano SPDA composites (36). Another natural powder, which is siwak powder, was added in different weight concentrations into HcAr. Siwak powder with average particle size of 75 µm at 5 wt% did not enormously impact the tensile and compressive strengths while the addition of this powder at 3%wt had minor influence on impact strength of the composites. The composite containing 7% siwak powder revealed a major reduction in tensile, compressive and impact strengths in comparison to the unreinforced polymer (37). Two types of modified cellulose were also incorporated into the HcAr where it was revealed that the addition of cellulose modified by 3-Methacryloxypropylmethyldimethoxysilane improved flexural strength while Triethoxy(octyl) silane did not display desirable results (38). Inhibition of *Candida albicans* and other fungi and bacteria on denture materials could have a major impact in alleviating denture stomatitis expansion. For this purpose, the effect of henna powder on *Candida albicans* was examined for reinforcing HcAr samples that were incubated in *Candida albicans* rich saliva at body temperature. Although *Candida albicans* proliferation was shown to control after incorporation of Yamani henna powder into HcAr, the authors suggested conducting further investigation regarding *Candida* effects on the toxicity, physical and pharmacokinetic characteristics of HcAr (39).

2.2 Nano-Nobel Metal Particles

Nano-nobel metals, particularly nano-silver (AgNPs) are of particular interest in a broad variety of applications due to their numerous properties such as high thermal and electrical conductivity, and chemical stability (40). These nanoparticles have the capability to exhibit notorious antimicrobial activities even at low concentrations against widespread types of viruses, fungi, and bacteria (41, 42). Nanoparticles size, synthesis methods, and shape have been observed to affect the antimicrobial efficiency of AgNPs (43-45). As a consequence of the abovementioned unique properties of AgNPs, besides their ability to selectively destroy cellular membranes, such nanomaterials can provide the cure and inhibition of numerous diseases and infections, particularly that are related to oral infections (46). A number of studies have been carried out using PMMA and AgNPs for the last few years to fabricate composites for dental applications. Alla and his colleagues investigated the impact of adding different percentages of AgNps on the flexural strength of Lucitone199, Trevlon, and TriplexHot heat-cure denture base materials. Incorporation of AgNps decreased the flexural strength of these materials in comparison with the unmodified polymers (47). AgNps were also added at 5 wt% to PMMA for the potential fabrication of removable prostheses. Incorporation of AgNps resulted in reducing the mean tensile strength of composite materials significantly (48) while swelling ratio was illustrated to increase with AgNP content (49). It was also indicated that flexure, compressive, and impact strength of the PMMA-AgNps composite prepared via photo polymerisation approach were improved in comparison with the pristine polymer (49, 50). However, Sodagar and his colleagues found that the influence of AgNps on flexural strength of PMMA relies on various factors including the type of acrylic and the mass fraction of nano particles. For instance, they noticed that incorporation of 0.05% AgNps into two types of PMMA namely, Rapid Repair and Selecta Plus significantly decreased its flexural strength for the former and increased it for the latter. While continuing rising AgNPs loading up to 0.2% increased flexural strength for Rapid Repair, this same ratio was less effective than 0.05% nano particles in improving the flexural strength for Selecta Plus (51). The antimicrobial behaviour of acrylic denture base containing various concentrations of AgNps and nanogold (AuNP) to *S. aureus*, *E. coli* and *C. albicans* was assessed. Broth Microdilution assay displayed that incorporation of both AuNPs and AgNps to PMMA could improve their antimicrobial activity (52). In another study, monodispersed citrate-capping AgNPs with a size of 20 nm and concentrations of 3 and 3.5 wt% were added into PMMA to study the antimicrobial behaviour of composites against *Candida albicans*. The topographical modification of PMMA and PMMA-AgNPs exhibited a reduction in surface roughness as was indicated by atomic force microscopy. The *Candida albicans* colonisation on PMMA surfaces was evaluated at day 1 and 2 with encouraging results on the reduction of yeast viability after AgNPs exposure (46). Acosta-Torres and his colleagues developed a novel biocompatible and antifungal PMMA-AgNps composite. MTT, enzyme-linked immunosorbent assay BrdU, and Comet assay showed that there was no harmful effect on cellular viability and cell proliferation and no genotoxic damage to mouse embryonic fibroblasts and Jurkat human lymphocyte cells. PMMA-AgNps composites were also revealed to significantly reduce adherence of *Candida albicans* (53). Various amount of AgNPs were also incorporated in PMMA to reduce denture stomatitis caused by *Candida glabrata*. Higher AgNPs concentrations led to decrease PMMA flexural strength although some other mechanical properties were improved. The nanocomposite with 0.05% of AgNPs had significant capacity to inhibit the biofilm formed on its surface as illustrated by the microbiological adhesion test against *Candida glabrata*. It was found that antimicrobial property was not linearly dependent of nanoparticles concentration and was affected by dispersion and distribution of nanoparticles in the matrix (54). Gold nano-particle (AuNP) can also be utilised to modified PMMA. The PMMA/AuNP composites were revealed to have increased thermal diffusivity and enhanced viscoelastic responses than unreinforced PMMA (55). Nanoparticles from another noble metal, which is platinum (PtN), were employed with PMMA to fabricate a potential novel antimicrobial composite. The formed composites were tested against *Streptococcus mutans and Streptococcus sobrinus*. PtN expressed significant bacterial anti-adherent impact when compared to bare PMMA in which these composites could have suitable antimicrobial activity with appropriate mechanical properties. For clinical application, the authors suggested carrying out further evaluating regarding long-term effect of this activity, biocompatibility and colour stability (56). It was shown that each of noble metals could have various influences on PMMA based composites. While silver and platinum could increase bending deflection, palladium was capable of improving bending strength and Vickers hardness (57).

2.3 Ceramic Fillers

A variety of ceramic fillers have been drawn considerable attention in recent years by many researchers to be potentially utilised in reinforcing PMMA denture base resin. It can be noticed from the literature that these materials may account for the higher portion among other reinforcement kinds. This may attribute to the broad range of properties that these fillers can provide. Some of these fillers can contribute, in one way or another, in fabricating high mechanical behaviour, anti-microbial and cost effective composites. It was revealed that mechanical properties such as Charpy impact strength, three-point bending and biaxial flexural strength of the conventional and high-impact (HI) denture base materials could be improved by incorporating silane coupling agents surface modified filler particles (58). TiO_2 nanoparticles with different percentages were blended with PMMA through a melt compounding process to form nanocomposites of PMMA plus TiO_2 for possible use in dental applications. Significant correlation between TiO_2 and PMMA matrix with a homogeneous distribution of TiO_2 in the polymer matrix was observed. The proper correlation between TiO_2 and the matrix resulted in improving the mechanical properties as confirmed by micro-indentation test, scratch test, and FESEM (59). It was noticed in another study that the degradation behaviour of PMMA was insignificant when small amounts of TiO_2 nanoparticles up to 3 wt% were added. The presence of TiO_2 in PMMA/TiO_2 nanocomposites led to enhancing the hardness, modulus, creep-recovery and relaxation behaviours of this composite. The antimicrobial activity of PMMA was also enhanced with TiO_2 due to decreasing bacterial adherence (60). A better distribution and a great increase in tensile strength were observed with 1wt% of TiO_2 NPs compared to other formula. Increase of TiO_2 content, on the other hand, reduced the tensile strength (61). In another study, 1% and 5% TiO_2

NPs was used to modify two types of PMMA. It was stated that flexural strength and microhardness considerably decreased and increased, respectively by increasing TiO_2 concentration (62). The incorporation of TiO_2 in the composite exhibited superb antibacterial characterisations compared to bare PMMA. It was suggested that the addition polyetheretherketone with TiO_2 at 1wt% of each can generate superb mechanical strength, great antibacterial activity, and low cytotoxic (63). Another promising reinforcing material that can be utilised to enhance some properties of PMMA is zirconium oxide nanoparticles (nano-ZrO_2). Gad and his colleagues investigated the influence of adding these fillers on the translucency and tensile strength of HcAr for removable prostheses. While the increasing of nano-ZrO_2 concentration led to improving the tensile strength significantly and proportionally, the translucency of the nanocomposites was reduced with such rising in the added nanoparticles (64). It was also displayed that the impact strength of nano-ZrO_2-reinforced specimens improved with a low loading of nano-ZrO_2 and reduced with rising nano-ZrO_2 mass fraction (65). Conversely, it was indicated that impact strength and surface hardness of the ZrO_2 reinforced specimens had relatively lesser values compared to the control specimens (Trevalon HI) (66). Addition of nano-ZrO_2 to PMMA increased the flexural strength, fracture toughness, and hardness of the composites (67). Furthermore, the addition of nano-ZnO particles increased also the flexural strength of the composites when compared to ZnO-free samples (68). Compressive strength was shown to be better for the PMMA/ nano-ZnO composite than that of the bear PMMA under identical conditions. Nonetheless, it was suggested conducting further investigations to prove that this improvement in compression behaviour was attributed to the used nanoparticles. Interestingly, it was found that this composite had no cytotoxicity effect on L929 cells (69). No significant deterioration in the mechanical properties of denture base was reported following the addition of nano-ZnO. The reduced growth of microorganisms was observed on ZnO-PMMA nanocomposites which could be explained by the increased hydrophilicity and hardness with absorbability (70). Generally, the flexural strength and modulus of the high impact (HI) heat-cured PMMA was reported to significantly enhance by the addition nano-ZrO_2 when compared to the neat PMMA. Nevertheless, this improvement was not the case concerning the fracture toughness of the composites, particularly at high concentrations of nano-ZrO_2 (71). Conventional water-bath curing method and microwave curing methods were utilised to produce composites from ZrO_2 nanoparticles and HI HcAr. The mean values of flexural strength and microhardness were shown to significantly increase for the samples prepared from HI HcAr (Trevalon Hi) plus 1% and 3% nano-ZrO_2. Better outcomes were achieved for the composite prepared using microwave curing than conventional method (72). The flexural strength of complete removable dentures fabricated from HI HcAr and nano-ZrO_2 at various concentrations was also examined. A mastication simulator was used to apply fatigue loading on the dentures and equivalent flexural strength was calculated with data obtained from bending tests with and without fatigue cyclic loading. Based on the obtained findings, the authors pointed out that the use of ZrO_2- PMMA in the fabrication of dentures did not lead to any noteworthy enhancement for clinical applications (73). In a different study, two types of additives namely, nano zinc oxide and nano-ZrO_2 were added to PMMA resin. While thermal conductivity and completion strength of the resulted composite were improved, roughness was decreased (74). Nano-ZrO_2 and micro-lignin particles were incorporated separately to cold cure PMMA with various volume fractions to produce complete prosthesis and partial denture base composites. The tensile modulus of elasticity, compressive strength and hardness improved with rising the volume fraction of these particles in PMMA. On the other hand, tensile strength, elongation and impact strength were intended to decrease. The incorporating of nano-ZrO_2 particles exhibited higher impact than that of micro-lignin particles in enhancing some of PMMA composites properties (75). ZrO_2 nanopowder can also be added with other materials to produce PMMA composites. Alqahtani studied the mechanical properties of composite materials fabricated from self-cured PMMA with hexagonal boron nitride (h-BN) and stabilized ZrO_2 (8Y ZrO_2) nanopowders. It was found that using h-BN nanopowder fillers led to higher bending strength and elastic modulus of the nanocomposite, whereas using 8Y ZrO_2 nanopowder resulted in improving hardness (76). ZrO_2 with other three kinds of nanoparticles namely, fly ash, fly dust, and aluminium were also added to cold cured PMMA for upper and lower prosthesis complete denture. Compressive strength and modulus under the effect of moisture and UV were risen with the addition of abovementioned nanoparticles (77). The effect of adding ZrO_2 into cold cured PMMA on *Candida albicans* adhesion was assessed after this composite was incubated at 37°C in artificial saliva containing *Candida albicans*. It was revealed that nano-ZrO_2 could have a major role in enhancing the performance of PMMA toward reducing *Candida albicans* adhesion to repaired PMMA denture bases and cold-cured removable prosthesis (78). Silica (SiO_2) nanoparticle have been added to PMMA to form nanocomposite materials for dental materials. PMMA-SiO_2 nanofiber synthesized by the electrospinning process was impregnated with methacrylate of BisGMA/TEGDMA (50/50 mass ratio). There were no statistically significant changes in flexural strength values between composite reinforced with PMMA-1wt% of SiO_2 nanofibers compare with PMMA nanofiber (79). A significant increase in flexural strength and modulus was achieved at the addition of SiO_2 nanoparticle. Although the addition of SiO_2 could negatively affect the formed composite biocompatibility, cytotoxic potential of adding SiO_2 nanoparticle toward L929 and MRC5 cell lines was on the acceptable level for specimens with up to 2% nano-SiO_2 (80). The sol-gel technique was utilised to formulate the PMMA/PS/SiO_2 composite membrane. The *in vitro* cytotoxicity studies conducted using MTT showed that the composites revealed high RAW macrophage cell viability; compared to PMMA and other composites, the composite with equal concentration of PMMA and PS demonstrated evidence of no inflammation (81). Muhammad and his colleagues blended PMMA with various concentrations of micro-fine amorphous SiO_2 and TiO_2 to produce composite materials for artificial teeth. In the presence of artificial saliva and under similar biological circumstances, the wear resistances were shown to enhance for these composites compared to commercial artificial teeth (82). Incorporation of TiO_2 and SiO_2 nanoparticles into PMMA can adversely affect the flexural strength of the produced composite. Higher flexural strengths for composites contained SiO_2 were demonstrated than those made of TiO_2 with the same concentration, yet the differences were not significant (83). SiO_2, TiO_2 and cerium oxide (CeO_2) were also added to PMMA at different concentrations. The fabricated composites had higher degree of conversion values and showed appropriate flexural strength and elasticity modulus even though CeO_2 could provide better overall properties of the dental PMMA composites (84). Some studies have investigated the capability of adding alumina (Al_2O_3) with several other materials in enhancing the mechanical properties of PMMA for potential use in dental applications. Samples of PMMA, PMMA/5HA and PMMA/10HA nanoparticles with various amounts of nano-Al_2O_3 were prepared. Not only the flexural properties and impact strength of hybrid nanocomposites were significantly enhanced in comparison with the bare PMMA, hybrid nanocomposites also had reduced shrinkage (85). Mixture of ZrO_2-Al_2O_3 nanoparticles was also added into HcAr, in which this addition led to highly significant increase in impact and transverse strength and significant enhancement in roughness of the composites. On the other hand, non-major improvement was observed regarding the hardness with reduction of adaptation for the composites compared to bare HcAr (86).

ZrO_2-SiO_2-Al_2O_3 filler reinforced PMMA was developed as novel composites. Sol-gel technique was utilised to produce this filler system while the composites were modified using chitosan and trimethoxypropilsilane (TMPS). The hardness values of chitosan modified composites were the higher followed by TMPS functionalised materials whereas the lowest values were for unmodified composites (87). In order to improve PMMA hardness, Hasratiningsih and his colleagues added various weight concentrations of ZrO_2-Al_2O_3-SiO_2 fillers to this resin at two calcination temperatures. The 13% reinforced ZrO_2-Al_2O_3-SiO_2 filler system that was calcined at 700°C exhibited higher Vickers hardness numbers compare to that was calcined at 550-700°C and the unreinforced polymer as well. Nevertheless, these values were not significantly different as they were verified statistically by ANOVA (88). A significant increase in the flexural strength of PMMA was observed after addition of 1% Al_2O_3 and TiO_2 nanoparticles in both heat- and auto-polymerised PMMA exceeding those modified with SiO_2. It was claimed that the mechanical properties of PMMA can be improved by the addition of this ratio of nanoparticles in which the addition of nanoparticles at higher percentages led to decreasing the flexural strength (89). Al_2O_3/yttrium stabilizer (YSZ) with nitrile-butadiene rubber (NBR) particles filled PMMA denture base material was evaluated for prosthodontics applications. NBR particles at 10 wt% was added to Al_2O_3/YSZ powders by ratio 50/50 with various loading prior to adding into PMMA matrix. Statistically, there were significant enhancements in the impact strength and fracture toughness for PMMA reinforced by NBR and Al_2O_3/YSZ. It was noticed that the impact and fracture strength of HcAr was enhanced after reinforcement with NBR particles and silane treated ceramic fillers while hardness was not remarkably improved (90, 91). Even though the above-mentioned studies showed promising results of using Al_2O_3, Rashahmadi and his colleagues found, by using nine multi-criteria decision-making procedures, that Al_2O_3 was the less appropriate reinforcement for PMMA denture compared to TiO_2 and SiO_2. They noticed that TiO_2 nanoparticles was the most appropriate material among these three nanoparticles for dentistry applications (92). Several sizes of h-BN nanopowder were also added into self-cured PMMA to make composites by using both hand and ultrasonic mixing methods. Ultrasonic mixing method was shown to provide better properties compared to hand mixing. While addition of h-BN nanoparticles to PMMA enhanced the hardness of PMMA, PMMA/nano h-BN composite fabricated using ultrasonic mixing method increased VH numbers to 300% and flexural strength values to 550% compared to the unmodified PMMA made by hand mixing (93). Among the added nanoparticles into PMMA, nanocarbon family such as nanodiamonds (NDs) is postulated to have excellent physical, chemical and antibacterial characterisations, including chemical stability, and biocompatibility (94, 95). Different concentrations of NDs were added to heat polymerised acrylic resin to form nanocomposite materials. The addition of 0.5% NDs to PMMA significantly increased and decreased its flexural strength and surface roughness, respectively. The addition of any NDs concentrations significantly reduced the impact strength of PMMA as statistical analysis of One-way ANOVA and Tukey's post-hoc demonstrated (96). The prevention of denture stomatitis could be achieved using composites of PMMA denture base and NDs. This was confirmed using two methods namely, slide count and direct culture test by examining NDs effects on *Candida albicans* adhesion. Statistical analyses for the resulted data revealed that the *Candida albicans* count. It was also revealed that surface roughness were significantly reduced after incorporation of NDs more than in the control group ($p<0.05$), whereas the contact angle was not significantly influenced (95). Calcium β-pyrophosphate (β-CPP) was also utilised for making PMMA/β-CPP composites for enamel regeneration and fixed-interim restorations. A statistically significant increase in flexural strength of the composites was found after adding β-CPP compared to the unreinforced PMMA. An obvious improvement of the mechanical properties and a slight improvement in the thermal stability of the composites were also observed as β-CPP content was increased (97). Some mechanical characteristics and the antifungal effects on *Candida albicans* and toxicity toward cells were studied for the composite made of PMMA plus silver bromide/cationic polymer nano-composite (AgBr/NPVP). The incorporation of AgBr/NPVP into the PMMA resin demonstrated strong and long-lasting antifungal impacts against *Candida albicans* of the composite. While the flexural strength and modulus were not significantly affected after adding these fillers, the resulted composite had low toxicity toward human dental pulp cells (98). PMMA was also reinforced by opaque dental porcelain powder for dental restoration applications. The composites were shown to have less water solubility, absorption and weight loss than that of PMMA. Based on the environmental and thermal tests, it was found that the properties of fabricated composites were consistent with the ISO standards for denture base materials (99).

3. Fibers-PMMA Composites

Interest in fiber-reinforced composite materials has increased remarkably in recent years, and this has been reflected in the use of fiber-reinforced composite products in almost all areas of life including biomedical applications. This may attribute to the unique properties that these materials possess in terms of their ability to form excellent mechanical properties, chemically stable, and inexpensive products when compared to others. For instance, fiber based PMMA composites could have better performance than the matrix and reinforcement alone where fibers improve the matrix strength, and the matrix provides protection to the fibers against the harsh oral environment. Due to the presence of a large number of fiber types with several sources, it can be easily altering the properties of the resulting composite material according to its intended use. Besides, it is also possible to improve the composite material behaviours by adjusting the fibers length, treatment, volume fraction, and the way the fibers are orientated and distributed within the matrix. In this part of the review, we will highlight the recent studies that have used fibers, whether natural or synthetic, in strengthening the PMMA resin for dentistry.

3.1 Natural Fibers-PMMA Composites

Natural fibers have widely been utilised due to their low cost, eco-friendly, biodegradability and biocompatibility make them favourable to be extensively used as a reinforcement phase in potential composites for dental field. Kenaf fibers was exploited by Theng and his colleagues to reinforce the PMMA resin. The authors found that flexural and compressive strengths of the composites were not improved when 1% and 2% of these fibers were added. They pointed out that inadequate surface treatment of fibers to provide adhesion between fibers and matrix could be the potential reason for the unchanged mechanical properties (100). In another research, chopped ramie fibers were used to fabricate PMMA-based composites. Due to good dispersion for 1.5 mm ramie fibers reinforced PMMA, flexural modulus of the composites increased considerably with fiber loading. Although the flexural modulus was increased more rapidly after incorporating 3.0mm fibers up to 4 v%, the modulus was reduced at higher ratios as a consequence of agglomeration. Flexural strength of short ramie fiber denture base PMMA declined because of the insufficient interfacial adhesion and it was claimed that the critical fiber length for the fiber in this kind of composites was 2.1 mm (101). John and his colleagues found that natural OPEFB fibers, especially OPEFB in cellulose form significantly improved the flexural strength and modulus of PMMA resin and therefore can be an alternative to commercially available synthetic fiber reinforced PMMA composites (102). Another natural fiber, which is siwak fiber, was chosen at various lengths and weight fractions to strengthen PMMA (103). Before being added into PMMA, the siwak fibers were treated with sodium hydroxide solution. The tensile strength, hardness, modulus and fracture toughness of composites improved with siwak fiber content, while the impact strength and elongation percentage decreased. Okeke and his colleagues (104) determined the characteristics of Hibiscus sabdariffa natural fiber as a reinforcement material for PMMA denture resin. Diameter, density, moisture content and absorption of Hibiscus sabdariffa fiber were observed to be within the range of other established reinforcing lingo-cellulosic fibers. The decreased hydrophilicity of sodium hydroxide treated fibers improved the thermal stability of the fiber. Furthermore, the adhesion between fiber and matrix was improved which resulted in improvement of composite mechanical properties. It was found that PMMA reinforced Hibiscus sabdariffa fibers produced higher flexural and impact strength mean values than unreinforced PMMA (105).

3.2 Synthetic Fiber-PMMA Composites

Despite the mentioned unique properties of the natural fibers, synthetic fibers can provide better mechanical behaviours for the dental-based composites. Polypropylene fibers with various weight percentages and aspect ratios were used to improve the flexural strength of PMMA based material. This composite showed improved flexural strength, particularly for the composite produced from 2.5 wt% of 6 mm long fiber (106). Furthermore, significant higher flexural strengths were obtained for hydrogen plasma-treated polypropylene fiber-reinforced composites due to enhancing the adhesion between PMMA matrix and the used fibers. Such improvement in flexural strength was specially obvious for the composite material contained 6 mm long fibers at 10 wt% (107). Untreated and oxygen plasma treated polypropylene fibers added to HcAr showed a highly significant increase in impact strength and surface hardness; nonetheless, transverse strength was not significantly changed. Besides, the composite containing oxygen plasma treated polypropylene fibers was revealed to have fracture resistance higher than that was fabricated from unmodified polypropylene fibers (108). It was also pointed out that the transverse strength and surface hardness of HcAr reinforced by randomly oriented 12mm long polypropylene fiber at concentration of 2% were higher than that was reinforced by 6mm long at the same concentration, or unreinforced HcAr (109). There were differences in flexural and impact strength, which could be either significant or highly significant between bear HcAr and the HcAr reinforced with various lengths of polyester fibers, polypropylene, and hybrid polyester and polypropylene. Therefore, the incorporation of hybrid polyester and polypropylene fibers could make HcAr more resistance to bend and break (110). Visible light cure fiber frame work was used as reinforcement material to increase acrylic denture base resistance to fracture. This kind of fibers was revealed to considerably increase the transverse and impact strength of the HcAr without affecting the surface hardness (111). Glass, polyaromatic polyamide and ultra-high molecular weight polyethylene fibers were added to HcAr. The flexural properties were significantly enhanced the addition following adding of glass fibers, polyethylene fibers, or aramid fibers to PMMA. Furthermore, hybrid fiber reinforced composite such as glass/ polyethylene fibers exhibited even highest toughness and the flexural strength making it potentially capable of preventing denture fractures clinically (112). It was found that the glass fibers could be more effective in improving impact strength of HcAr even better than metal wire (113). E-glass fiber was also employed as chopped strands of 3 mm size with various weight concentrations for reinforcing HcAr. The obtained data illustrated that addition of 1% and 1.5% E-glass fiber improved the impact strength, transverse strength, and modulus of elasticity of PMMA. Even so, the addition of 1% E-glass fiber could create favouring balance of these properties of the resin (114). The mean flexural strength of the samples repaired with impregnated fiberglass into PMMA was higher than that of HcAr and self-curing PMMA. It was pointed out that the occurrence of fractures was due to the lower flexural strength of the auto-polymerising PMMA compared to that of the HcAr (115). PMMA reinforced with glass fibers demonstrated the highest flexural strength followed by butadiene styrene reinforced PMMA, and the least strength was detected in PMMA (116). The use of silanized allows obtaining higher flexural strength and these silanized fibers can act as crack stopper (117). The reinforcing effects of glass fiber mesh with various loadings and structures were compared with that of metal mesh in complete dentures. It was observed that the effectivity of glass fiber mesh was more than metal mesh, and the structure of the glass fiber mesh was as not important as its content (118). Measuring and comparing the fracture strength of HcAr reinforced with fibers of glass, polyaramid, and nylon were performed by Kumar and his colleagues They concluded that embedding of 4w% glass fibers in loose form enormously rose the transverse strength of PMMA, while the same content of randomly distributed polyaramid fiber considerably enhanced the transverse strength of denture base PMMA (119).

4. Fiber-Particle Hybrid Reinforcement

The presence of both ceramic particles and fibers could promote creating balance environment in terms of appropriate thermal, mechanical and biological properties of the hybrid nanocomposite. For this reason, the composites of PMMA plus 3-Methacryloxypropyltrimethoxysilane modified nano-ZrO_2 and electrospun polystyrene (PS) fibers were developed. Increasing the concentration of modified nanoparticles resulted in improving microhardness, whereas the used fibers reduced hardness and enhanced toughness of the composites. Moreover, absorbed energy for the composites was shown to rise following the incorporation of PS fibers (120). The hybrid reinforcement effects of nano-ZrO2 and glass fibers at different concentrations on the flexural and impact strengths of heat polymerised PMMA denture base composites was evaluated for potential removable prosthesis use. Significant improvements in the flexural and impact strengths for the PMMA-nano-ZrO_2/GF composites were found when compared to those of pristine PMMA (121). Hamad studied the effect of adding 0.5 %, 1 % and 1.5 % volume fractions of nano-SiO_2 particles and 3% volume fraction of random woven fiber glass on some mechanical properties of prosthesis complete PMMA composite materials. The flexural modulus, flexural strength, shear stress and impact strength were increased with the rise of the volume fraction of nano-SiO_2 particles in the composites; the values of flexural strain decreased (122). A novel botryoidal PMMA microsphere-grafted aramid fiber composite was developed to be potentially used in the field of restorative materials. The addition of a microsphere-grafted aramid fiber to PMMA reduce the aesthetic problem of the aramid fiber, simultaneously enhanced the mechanical properties with acceptable *in vitro* safety cytotoxic of the fabricated composite (123). Several loadings of silanized nano-ZrO_2 and aluminium borate whiskers (ABWs) were mixed with PMMA to formulate ZrO_2-ABW/PMMA composites. The mechanical behaviours such as flexural strength and hardness of silanized ZrO_2-ABW/PMMA composites were significantly improved (124). Hybrid addition of nano-ZrO_2/ glass fibers to PMMA improved the tensile strength and the highest value was noticed with the incorporation of 50% nano-ZrO2/ 50% glass fibers. On the other hand, weak adhesion and pull out of fibers from the matrix were detected in high glass fibers loading (125).

5. Nanotubes-PMMA Composites

Another fascinating kind of materials whose its use has increased dramatically in the last few years is nanotubes. These nanomaterials provide unique properties resulting from providing a high surface area, which allows improving the composite materials characteristics that they are made by distributing them to the loads and stresses imposed on the composite material, thus protecting the matrix from breakage and failure. Furthermore, the availability of the high surface area of these materials may increase the interaction between the matrix phase and the reinforcing material in the composites. Moreover, the presence of porous structures for most of these materials may increase the opportunity of penetration of the base material and thus increase the interfacial connection between the components of the composite material, especially if the nanotube contains functional groups that can interact with other groups present in the monomer. Interestingly, some studies revealed that the composites fabricated from nanotubes could have better properties than that formed with nanoparticles of the same material. Yu and his colleagues found that ZrO_2 nanotubes was demonstrated to have a better reinforcement effect than ZrO_2 nanoparticles when they added to PMMA. Furthermore, untreated ZrO_2 nanotubes displayed more effective reinforcement than silanzed treatment ZrO_2 nanotubes in which the composite produced from 2.0 wt% untreated ZrO_2 nanotubes revealed higher flexural strength (126). Flexural strength and hardness of specimens modified with both 2.5 and 5 wt% TiO_2 nanotubes were also significantly higher than those of unmodified PMMA. Although fracture toughness of samples reinforced with 2.5 illustrated different behaviour to flexural strength and hardness, fracture toughness at 5 wt% TiO_2 nanotubes was higher than neat PMMA (127). Another well-known nano-reinforcement material is carbon nanotubes (CNT). It has astonishing modulus of elastic, tensile strength, and high electrical conductivity in addition to its lightweight and high aspect ratio. Such properties make it extremely desirable fillers for polymers reinforcement in a wide range of applications including automobiles, airplanes and antistatic packaging (128, 129). Various types of carbon nanostructures, such as single walled carbon nanotubes (SWCNTs) and multiwalled carbon nanotubes (MWCNTs) are available. Both of them have commonly been utilised in several applications, including protein, gene, and drug delivery (130-132). The flexural and impact strength of three commercially available denture base resins after reinforcing them with SWCNTs were investigated. The flexural strength of each material also increases gradually with increasing percentage of SWCNTs addition (133). A significant increase in impact strength and transverse strength were noticed when carbon nanotubes were added at 1wt%, compared to pristine PMMA while hardness was decreased due to this addition (134). It was also found that adding 1.5% of SWCNTs considerably increased the impact and transverse strength of HcAr, but considerably reduced its hardness (135). MWCNTs at 0.5 and 1 wt% were shown to significantly improve flexural strength and resilience for this composite higher than unreinforced PMMA and the composited formed by using 2 wt% MWCNTs. The interfacial bonding between MWCNTs and PMMA matrix was not strong enough as the Raman data and dynamic loading findings showed. MWCNT undesirably affected the fatigue resistance of MWCNT-PMMA with even more deteriorating at higher concentrations of MWCNTs (136). The mechanical properties of Luciton-199® were shown to enhance by the addition of MWCNTs at 0.5 and 1 wt% to PMMA. The concentration and dispersion of the MWCNTs added into PMMA matrix were essential factors in determine the range of MWCNT reinforcement efficiency (137). A highly significant increase in tensile and transverse strength, hardness and important decline in water sorption and solubility were observed for the composites prepared from HcAr with various weight concentrations of both SiO_2 and CNT compared to unreinforced HcAr (138). The non-toxicity, biocompatible, functionality and reasonable price availability of halloysite nanotubes (HNTs) make them utilised for various applications such as fabrication of nanocomposites, drug delivery and bone regeneration (139-141). Incorporation of lower concentration of HNTs at 0.3 wt% into PMMA produced a significant increase in hardness; nonetheless, hardness significantly decreased for the formed composites at higher concentrations. Regarding the flexural strength and modulus values, their increases were not significant compared to that of unmodified polymer (142). Despite the advantages mentioned for the nanotubes, it could be alleged that there has been less research done on the use of these materials in dentistry when compared to other materials. This may be related to heavy cost for some of these materials and the relative difficulty in their handling, and surface treatment and functionalisation as well as to their toxicity *in vivo*. Therefore, developing these materials by reducing the limitations that hinder their use in medical applications, or finding other types with better characterisations may open the door to major development in the biomedical fields and the dental industry in particular.

6. Conclusions and Recommendations

Although PMMA has amply been utilised for a long time in dental applications, some limitations such as the low resistance to impact and fracture during practice, strength, and viscoelastic behaviour are still present in PMMA denture base material. This prompted many researchers to conduct many studies for the purpose of improving the PMMA properties and performance in this field. Efforts have been made for developing PMMA composite materials reinforced with materials of natural and synthetic sources in the form of particles, fibers and nanotubes. Examples of the particles that have extensively been utilised for reinforcing PMMA are noble metals including silver, gold, and platinum. In addition to this kind of particles, ceramic materials such as metal oxides, minerals and diamonds have commonly been used. These materials can provide improved mechanical characteristics, excellent aesthetic appearance, and long-lasting antifungal and bacterial properties. Regarding fiber reinforced dental composites, the performance of these materials could be tuned by a number of factors namely, fibers type, loading, length, and orientation. The PMMA/fiber composites that have recently been fabricated for dental industry can be natural, synthetic or natural/synthetic hybrid. The interest in natural fibers composites has been increased recently especially those have been fabricated from plant-based fibers because of its favourable biocompatibility, easy fabrication, environmentally friendly and cost effectiveness. On the other hand, synthetic fibers such as glass and polypropylene fibers have played an important role in forming composites with good flexural strength and hardness, with the possibility of improving their properties even more by increasing the interfacial adhesion and interconnection between the fiber and the matrix through the surface treatment of the fibers. Finally, the studies that have investigated the use of nanotubes in enhancing the properties of dental composite materials were addressed. The advantages of nanotubes such as the large surface area, their superb mechanical behaviours and their light weight can positively affect the final product performance. Despite the noticeable improvements in the performance denture PMMA composites, some of the examined composites are still unable to achieve the desired ideal performance for dental technology; therefore, further investigation could be required in this respect. Furthermore, some studies need to be conducted *in vivo* for the final decision about their uses in dentistry.

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