Original Research Paper

Volume - 12 | Issue - 03 | March - 2022 | PRINT ISSN No. 2249 - 555X | DOI : 10.36106/ijar

Dental Science

STRENGTHENING PROPERTIES OF PMMA WITH FILLERS FROM MACRO TO NANO SPECTRUM: A REVIEW

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ABSTRACT PMMA, the most largely used material in acrylic denture fabrication, has many favourable characteristics, but also several disadvantages, regarding its physical, chemical, biological and mechanical properties. Extensive research has been done, in order to improve PMMA mechanical properties, by changes in the chemical structure, material reinforcement using fibres or fillers of different types, sizes, shapes, in different concentrations, or by obtaining hybrid materials. With the advent in extensive nano research newer and better competent reinforcements are shown light of the day. Hence, the drawbacks shown by PMMA can be put back with these newer materials. Various scientific reviews, in-vitro studies, a few in-vivo studies, patient feedback were taken and here is a review wherein articles ranging from the year 1974 to 2021 were referred for the reinforcement of PMMA with a wide range of materials as it is said that there is no particular ideal material, a mix and match of all strengthens the basic PMMA. Specially the advent in Nano world with the carbon family is the newest of its kind.

KEYWORDS : Reinforcement, Denture Base, PMMA, Nano Fillers, Nano particles, Zirconia, Graphene, Carbon Fillers, Poly Methyl Methacrylate

INTRODUCTION

The presence of the denture in the oral cavity exposes it to a variety of forces during mastication, and also the interaction with the oral fluids and different beverages & food items. Poly Methyl Methacrylate is the most widely used denture base material till date, Although exhibiting many favourable properties, it cannot be considered an ideal material. Complete dentures have been fabricated from various materials namely wood, bone, rubber, plastics and what not; as its history is dated back to 700 BC . Several attempts have been done in time at improving the properties of acrylic materials used for denture fabrication. Keeping in mind the various mechanical, chemical, physiologic properties and processing the most ideal material is always the ultimate goal. Firstly, in the year 1937 Walter Wright and Vernom Brothers used PMMA as a denture base material keeping in mind its many properties and characteristics such as ease in processing, stability in oral environment, lightweight, aesthetic value, low cost, low water absorption and it can be easily repaired and changed.[1,2]Uptill the 1940's PMMA became the most promising and prevalent denture base material. It was later found that certain drawbacks such as low flexural strength, residual monomer allergy, porosity, polymerisation shrinkage, poor conductor of heat, poor adhesion to metal, high coefficient of thermal expansion, low impact strength, crazing, warpage all of these lead to experimenting and creating newer or chemically changed materials which could avoid these drawbacks in the most possible manner. [3,4] With the passing years and today in the decade of millennials extensive research has been done, in order to improve PMMA mechanical properties, by changing its chemical structure, material reinforcement using fibres or fillers of different types, sizes, shapes, in different concentrations, or by obtaining hybrid materials.[5] The stream of Nanosciences is vast and research done is quite little hence newer opportunities are created with each passing year. With various advents in the field of Macro to Nano science opens doors to materials which can enhance PMMA properties to an innovative approach. Thus this article in particular reflects and reviews upon the various fillers, reinforcements, particles which can be incorporated in PMMA to enhance its properties. This review paper discusses the many suggested materials from the very macro to nano level as to improve the most widely used PMMA.

Need for Reinforcement:

Even though PMMA was considered to be the ideal material[2] for denture base resin it has its own drawbacks hence the material needs enhancement in the form of reinforcements at different levels for changes in Physical, Mechanical as well as Chemical properties[4]:

	Drawbacks of PMMA
1.	Low flexural strength
2.	Residual monomer allergy
3.	Low Modulus of Elasticity
4.	Poor conductor of heat
5.	Polymerisation Shrinkage
6.	Warpage
7.	Crazing & Porosity

Hence after keeping in mind these properties newer materials and fillers were added and tried to overcome certain drawbacks and to bring about changes with these reinforcements. Several attempts have been done in time at improving the properties of acrylic materials used for denture fabrication.

The properties of PMMA so far have been modified by various methods such as[5]:

i) Chemical modification
ii) PMMA reinforcement at Macro & Micro Scale
iii) PMMA reinforcement at Nano scale
iv) Hybrid Reinforcement

Chemical Modification

Enhancement of PMMA thermal, chemical, physical and mechanical properties is potrayed by cross-linking, which describes polymer modification by interlinking polymer chains through ionic or covalent bonds; indulging the free movement of polymer chains is impeded and they just slide against each other. [6] Many authors tried improving PMMA properties using different polymerization techniques, like microwaves, visible light or autoclave polymerization,[7,8] to influence the amount of residual monomer; a higher temperature, for example, can increase the conversion rate of the monomer, reducing its residual content and, consequently, improving the mechanical properties.[7]

Thus different chemical modification methods were tried to further reduce the drawback structure of PMMA.

Let us discuss about Macro, Micro, Nano and Hybrid materials which are used for reinforcements:

Table 1: Enlisted are the fillers, nanofillers, hybrids which have been used for PMMA reinforcement.

Additives	Fillers	Macro/Micro Modification (µm)	Nano Particles (nm)
Metal Oxides	Alumina	Al_2O_3	Al_2O_3 NPs, Silane treated Al_2O_3
	Zirconia	ZrO_2	ZrO_2 NPs, Silane treated ZrO_2 NPs
	Titanium	TiO_2	TiO_2 NPs
Noble Metals	Silver	Ag	Ag NPs
	Gold		Au NPs
	Platinum		Pt NPs
	Palladium		Pd NPs
Silica Based	Glass Flake Mica Surface Treated Silica	SiO_2	Nanoclay
Fiber	Glass Fiber	Silanized Glass Fiber	
	Aramid		
	Nylon	Nylon Polyamide	
	Polyethylene		
	Polypropylene	Polypropylene Fiber	
Carbon	Hydroxyapatite		
	Graphene	GO	GO NPs
			Carbon Nanotubes
			Carbon Black
			Carbon Nanofillers
Miscellaneous	Metal Wires		
	Metal Mesh/Plates		
	Rubber		
Hybrid Reinforcements	Hybrid Fibers Metal Oxides and Ceramics Fibers and Fillers		Carbon Fiber and Graphene Graphene & CNTs Graphene & Silver Nanoparticles

From the Macro to the Nano Spectrum:

Initially, Metal wires were used to enhance few of the physical and mechanical properties of resins, such as flexural strength and tensile strength[9], whereas perforated metal plates determined a higher flexural strength and flexural modulus [10]. Even, many types of fibers were used for reinforcement in the previous years but it was discovered later that only an adequate adhesion between fibres and the composite matrix permits stress transfer from the matrix to the fibers, otherwise the stress may concentrate around the fibres, weakening the material.[11]

Studies have been conducted on the use of fillers such as Metal Oxides, Noble Metals, Silica based glass flakes and mica to reinforce denture base resin, which has significantly improved the properties of PMMA. Poly (methylmethacrylate) (PMMA) reinforced with metal oxides resulted in the improvement of the physical and mechanical properties of the denture base material. Patient's sensation of hot and cold stimuli was also improved. [12-14] Nowadays, it has been suggested and also practiced to incorporate nano-fillers of the same macro-fillers to improve PMMA properties. The high surface area, and homogenous nano-fillers distribution are the main properties which enhance the thermal properties of PMMA.[14,15] The properties of resin reinforced by nano-fillers depend on the size, shape, type, and concentration of the added particles.[15]

Metal Oxides

Alumina (Al_2O_3)

Flexural strength, impact strength, tensile strength, compressive strength, surface hardness, and thermal conductivity of the resin these were greatly increased.[16-23] Decrease in warpage was also a factor enhanced but certain studies found that alumina decreases PMMA's impact and tensile strength.[24]

Equal stress distribution all over the new reinforced material wasn't noticed and metal was poorly joined to polymer, weakening the resin hence methods were suggested to improve the bond by sandblasting, silanization and metal adhesive resins. A significant increase in the wear resistance was observed by treating aluminium with silane coupling agent.[25,26]

Disadvantage: Discolouration of resin (Cannot be used in visible areas).

Zirconia (ZrO_2)

Multiples studies have proved a prominent increase in flexural strength, impact strength, tensile strength, fracture toughness and thermal conductivity by adding Zirconia.[24-33]

Disadvantage: Due to clustering of particles within the resin, it weakened the material and slight decrease in flexural strength was noted.[34]

ZrO_2NPs

Adding Nano particles improves the mechanical properties along with increasing impact strength, flexural strength, compressive strength, fatigue strength, fracture toughness and hardness. It also potrays antifungal properties hence has a preventive role.[34-41] Also, adding Nano particles showed no color change.[42] Significant decrease in Water sorption and solubility of PMMA was the most important factor found.[13]

Silanized zirconia NPs improved the tensile strength, fatigue strength, hardness of PMMA. While, its porosity, water sorption, and solubility decreased.[42-45]

Titanium (TiO_2)

Titanium provided the same enhancement in properties such as zirconia impact strength, flexural strength, compressive strength, fatigue strength, fracture toughness, low water sorption and hardness. In addition thermal conductivity, was better with titanium.[13,20,29,46-48]

Disadvantage : Here, too Particle clustering was observed which lead to decreased flexural strength.[46]

TiO_2NPs

Decreased E-modulus, decrease in thermal expansion coefficient and contraction, Thermal Stability were all positive factors with the effect on mechanical properties but a reduction in flexural strength and toughness was observed.[48]

Adhesion between resin matrix and filler particles is of utmost importance hence comes the role of titanium coupling agent.[49] Silanized Nano particles improve transverse strength, impact strength and surface hardness also decreasing the water sorption. [50]

Noble Metals

Silver Nanoparticles (Ag)

Incorporation of Ag NPs is beneficial for immune-compromised patients as studies have shown antifungal properties [51-54], especially at high concentrations[55], and acted like a latent antifungal material with low-releasing Ag+.[56] However, Wady et al showed that it did not affect Candida Albicans and a biofilm accumulation.[57]

It increased flexural strength, fatigue strength and improved thermal conductivity. Comprehensive strength can be increased by silane-treated silver particles. Due to the improvement of thermal conductivity and compressive strength it is recommended to be used in the palatal area of maxillary dentures.[58,59]

Disadvantage: Poor Colour Stability, Incorporating 2% Ag decreases tensile strength.[60,61]

Nanoparticles – Gold (Au), Platinum (Pt), Palladium (Pd)

Though the studies on these noble metals is quite limited improved flexural strength and thermal conductivity double the normal value is noted.[62,63] Platinum NPs was found to have antimicrobial effect as well as it increases PMMA's bending deflection.[64] Here gold and palladium improved Vickers hardness while platinum decreased it.[65]

Silica-Based Fillers

Glass flakes

It's incorporation to acrylic resin improved its fracture toughness, and

silane coupling agent used resulted in further improvement of the acrylic resin properties.[29]

Mica

A group of lamellar silicate minerals and these are characterized by their high aspect ratio. They improved the mechanical, thermal, as well as dimensional properties of PMMA.[66] It particularly increased the hardness of acrylic resin, but its flexural strength was reduced because of weak bond of mica with the acrylic resin.[67]

Nanoclay

A material which is used to enhance the properties of acrylic polymers such as thermal conductivity, while flexural strength was decreased.[68]

Fibers

Different types of fibers are used such as nylon, polyethylene, polyamide fiber and glass fiber showing improvement in flexural and impact strength. Glass fiber being of utmost importance due to the aesthetics and mechanical properties.[69-76] Preimpregnated and silane treated glass fiber also increased the flexural strength[77] and impact strength[78,79] of acrylic resin. nanopigmented PMMA showed reduced porosity and Candida albicans adherence.

Flexural Strength, impact strength, Vickers hardness were significantly increased with glass fiber.[71-75] Incorporating silanized polypropylene fiber in PMMA resin significantly improved its transverse, tensile, and impact strengths, but its wear resistance was highly decreased. Polyamide fiber includes both Nylon and Aramid fiber. Denture base resin reinforced with aramid fiber was found biocompatible, and its flexural strength and flexural modulus were increased.[80-82]

A recent study found that the position of glass fiber within the denture base affects its flexural properties. Improvement of flexural strength, toughness, and flexural modulus was obtained from placement of glass fiber close to the surface of the denture base on its tensile stress side. When glass fiber was placed in neutral stress area, only flexural toughness was improved, and when placed in the compressive side, surface flexural modulus was increased.[76] Polypropylene fiber to PMMA significantly increase its impact strength, and a further increase was observed with fiber surface treatment.[79] The highest impact strength was obtained with polypropylene fibers treated with plasma, which can be used to strengthen acrylic resin and reduce fracturing.[82-84] Incorporating silanized polypropylene fiber in PMMA resin significantly improved its transverse, tensile, and impact strengths, but its wear resistance was highly decreased.[85,86]

Disadvantages: Polyamide- hardness of the resin decreased with increasing fiber concentration. The yellow colour is also considered unpleasant.[82,83]

The advent in the nano technological word has brought about a change immensely, development of new nano materials with properties different from those of the original material, due to their high active surface, compared to their smaller size.

Carbon Family Nano Fillers

Uptill, 5 years back carbon family fillers were not commonly used but with the introduction of graphene and carbon nanotubes in the dental material world changed the scenario Carbon nanotubes incorporated in PMMA determined better mechanical and electrical properties, and also a higher thermal conductivity.[87] Studies showed that nanofillers addition is more efficient than that of microfillers in improving the properties.[88] Surface functionalization of CNTs is an effective method to improve their dispersion, as a result of the strong interfacial interactions. CNT sponge, or CNT foam can find applicability in modern composites, due to its high porosity and very low density.[89-91]

Graphene is an easy-to-obtain and cheap material which has exceptional thermal, mechanical, chemical and electrical properties[92], was combined with various metals, carbon materials or polymer materials, inducing improved properties of the obtained composites . Reinforcement of PMMA using graphene or graphene oxide leads to a higher impact strength [93]. The higher functionalization of GO, compared to G, determines better results, as it facilitates the interfacial interactions between PMMA and GO. Side effects – like the rise of the residual monomer amount at higher concentrations of G and GO, and a delay in the polymerization reaction because of graphene interference – are also possible.[94]

Hybrid Reinforcements

Hybrid reinforcement, obtained by combining different types of additives, showed significant improvements in the mechanical properties of acrylic resins.[95] PMMA with carbon fiber coated by graphene oxide showed enhanced mechanical performances than the PMMA carbon fiber composite.[96] A graphene and CNTs hybrid material can also be an efficient solution for multifunctional interfacial nano reinforcement, improving the mechanical and electrical properties in carbon fiber and epoxy composites [97]. A combination of graphene oxide and silver nanoparticles showed favorable effects on several mechanical, chemical and physical properties of a PMMA denture resin.[98,99]

Vallittu first suggested different hybrid reinforcements in the year 1997, different combinations can be used such as metal oxides, ceramic material with fibers and nano fillers.[100-102] combination of fibers and other fillers increased impact strength, hardness, surface roughness, and thermal conductivity, as well as compressive and fatigue strengths.[108,109] Hybrid materials used are: Al2O3 + ZrO_2 , ZrO_2-TiO_2, Glass fiber + polyethylene fibers, Polyester fiber reinforced PMMA + (clay, glass powder, SiO_2, or ZrO_2).Al_2O_3 + plasma-treated polypropylene fiber nHA particles, micro-zirconia, glass fiber, and Kevlar fiber (PMMA-nHA and glass fiber), (PMMA-ZrO_2 and glass fiber), (PMMA-nHA and Kevlar fiber), (PMMA-ZrO^2 and Kevlar fiber).[100-110]

Summary

The denture in the oral cavity is exposed to a variety of forces during mastication, and also to the interaction with the oral fluids and different beverages. PMMA, the most widely used material for a long time now in denture fabrication, exhibits majority favorable properties, cannot be considered an ideal material. Several attempts have been done in time at improving the properties of acrylic materials used for denture fabrication. A higher impact, flexural, compression or tensile strength were obtained by structural changes in PMMA, incorporation of different additives such as wires, platelets, micro or nano fibers and fillers, or hybrid materials synthesis. The new era of nanosciences opens new perspectives in the improvement of PMMA based resins used in dentistry and in many other domains and hence much more research can be done which might give a material which is next to ideal or ideal in itself. Till then, the reinforcements strengthens the properties of PMMA.

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