Effect of Alumina Addition on Properties of Poly -methyl methacrylate - A Comprehensive Review

Pooja Arora[#1], S P Singh[#2], Vipin Arora[#3]

[#1,2]Department of Prosthodontics, Dental materials and Oral Implantology, Subharti University, Meerut, India

[#3]Department of Conservative Dentistry and Endodontics, Subharti University, Meerut, India

***Abstract*—** Polymethyl methacrylate (PMMA) is one of the most widely used materials in prosthetic dentistry. Since it was introduced to dentistry, it has been successfully used in construction of denture bases. Acrylic resins have been successful as denture bases because of their ease of processing, low cost, light weight, and colour matching ability,however, acrylic resin denture base materials are low in strength, brittle, and low in thermal conductivity.

Various methods to strengthen acrylic resin have been suggested. However, reinforcement methods should not have adverse effects on the other properties of denture materials. Although often overlooked, the heat transfer characteristics of the denture base material may be an important factor in determining patient satisfaction. In literature, few attempts have been made to develop acrylic resins that not only possess improved mechanical properties but also an overall improvement in the physical properties like thermal diffusivity, hardness, surface roughness and water sorption without negatively affecting each other.

The incorporation of ceramic particles in various dental materials has been studied and found to be biocompatible, and it also improves mechanical properties. In addition, the white colour of the ceramic powder is not expected to compromise aesthetic appearances. Aluminium oxide (Al_2O_3), commonly referred to as alumina, possesses strong ionic interatomic bonding, giving rise to its desirable material characteristics. Its high hardness, excellent dielectric properties, refractoriness, and good thermal properties make it the material of choice for a wide range of applications.

The aim of the present article is to review the effect of addition of alumina on various physical, mechanical and thermal properties of acrylic resins.

***Keywords*—**Alumina, acrylic, ceramic, denturebase, Polymethyl- Methacrylate, reinforcement, resin.

I. Introduction

Polymethyl methacrylate (PMMA) is one of the most widely used materials in prosthetic dentistry. Since it was introduced to dentistry, it has been successfully used in the construction of denture bases. Acrylic resins and rubber reinforced acrylic polymers represent approximately 95% of the denture base materials used in prosthodontics[1].Acrylic resins have been successful as denture bases because of their ease of processing, low cost, light weight, and colour matching ability [2],however, acrylic resin denture base materials are low in strength, brittle, and low in thermal conductivity [3,4].

Flexural strength of denture base resin is considered primary mode of clinical failure. A study by Johnston et al showed that 68% of acrylic resin dentures break within a few years after fabrication [5]. This occurs when the denture is accidentally dropped on a hard surface or fractures when subjected to high mastication forces.

Various methods to strengthen acrylic resin have been suggested. These include chemical modification to prepare high impact resin and mechanical reinforcement with glass fibers, sapphire whiskers, aramid fibres, carbon fibres, metal wires and plates, metal powder fillers (Silver, copper, and aluminium particles), nylon polyethylene fibres and zirconia. The primary problem with using metal wire is poor adhesion between the wire and resin, which leads to insignificant enhancement of mechanical properties. Although metal plates increase the strength, they may be expensive and prone to corrosion [6-9]. Modifications of the chemical structure by adding crosslinking agents or copolymerization with rubber results in significant increase in impact strength. However, stiffness, fatigue resistance, and transverse strength are reduced [10-12]. Mechanical reinforcement of acrylics has also been attempted through the inclusion of fibers and metal inserts [13-17].Although the inclusion of the fibers produced encouraging results, this method has various problems including tissue irritation, increased production time, difficulties in handling, the need for precise orientation, and placement or bonding of the fibers within the resin [18]. In the case of metal inserts, failure due to stress concentration around the embedded inserts has been reported [17,19-20]. Also, metal-reinforced dentures were found to be unesthetic.

The incorporation of ceramic particles in various dental materials has been studied and found to be biocompatible, and it also improves mechanical properties [20-28]. In addition, the white colour of the ceramic powder is not expected to compromise aesthetic appearances [20,23,29,30].However, reinforcement methods should not have adverse effects on the other properties of denture materials.

The roughness of acrylic resin surfaces is a critical property because surface irregularities increase the likelihood of microorganisms remaining on the denture surface after the prosthesis is cleaned [31,32].

Another property that can influence the surface characteristics of acrylic resins is the hardness, which indicates the ease of finishing a material and its resistance to in-service scratching during cleaning procedures [33]. The absorption of water by acrylic resins is a phenomenon of considerable importance. Acrylic resins absorb water slowly over a period of time, primarily because of the polar properties of the resin molecules. High equilibrium uptake of water can soften an acrylic resin because the absorbed water can act as a plasticizer of acrylic and reduce the strength of the material [34]. According to Anusavice,it has been estimated that for each 1% increase in weight produced by water sorption, acrylic resin expands 0.23% linearly [35].

Although often overlooked, the heat transfer characteristics of the denture base material may be an important factor in determining patient satisfaction. For example, it has been established that food temperature significantly affects the perception of taste [36-39].Because the process of eating consists of frequent and abrupt changes in the temperature of food, thermal characteristics of the denture base material can become important factors affecting the gustatory response, for example, chemical perception of taste, smell, textural perception, and temperature.

In a study by Kapur KK and Fischer EE(1981), thermal conductivity, or rather, the transient thermal conductivity (thermal diffusivity) of the denture base material was found to have an important effect on parotid secretions, which have been shown to be a good approximation of gustatory sensitivity [40,41]. In edentulous patients wearing full dentures, the palate is partially covered by the denture base; consequently, the ability to sense transient temperature changes at the palate may be affected by the thermal characteristics of the denture base material.

The acrylic polymer most commonly used for denture bases, polymethyl methacrylate (PMMA), has a thermal conductivity of approximately 0.2 *W/minl°K*, which is approximately three orders of magnitude less than most metals[3].It is therefore not surprising that thermal conductivity has been one of the properties of acrylic resins most often associated with their replacement with metal as a denture base material (for example, gold and chromium cobalt alloys) [1,42-44]; however, the use of metal as a denture base material has several disadvantages including increased weight of the denture, difficulty with tissue replacement in cases where substantial loss of bone has occurred, difficulty restoring denture borders within physiologic boundaries, difficulty with the relining process, esthetics, and high cost.

Because of the disadvantages associated with metallic denture base materials, exploring the development of acrylic-based materials with improved thermal diffusivity is of interest. One approach to improving thermal diffusivity of denture base acrylic resins is to introduce a more thermally conducting phase within the insulating acrylic resin matrix, which creates a composite denture base material.

Researchers have attempted to add particles of a conducting material, namely metal, to the powder or liquid resin and polymerize according to established procedures, increasing thermal conductivity modestly [45], however, to significantly increase thermal conductivity with this approach requires high metal powder loading (>25%),which alters some of the beneficial characteristics of acrylic resins such as increased density due to the presence of large amounts of metal, lower tensile strength and toughness, and diminished esthetic appearance due to the metal. In literature,few attempts have been made to develop acrylic resins that not only possess improved mechanical properties but also an overall improvement in the physical properties like thermal diffusivity, water sorption without negatively affecting each other.

Aluminum oxide (Al_2O_3), commonly referred to as alumina, possesses strong ionic interatomic bonding, giving rise to its desirable material characteristics. It can exist in several crystalline phases, which all revert to the most stable hexagonal alpha phase at elevated temperatures. This is the phase of particular interest for structural applications. Alpha phase alumina is the strongest and stiffest of the oxide ceramics. Its high hardness, excellent dielectric properties, refractoriness, and good thermal properties make it the material of choice for a wide range of applications [46-61].The aim of the present article is to review the effect of addition of alumina on various physical, mechanical and thermal properties of acrylic resins.

II. Effect of Addition of Aluminium Oxide on Various Properties of PMMA

Flexural strength

Alumina addition leads to both increased and decreased flexural strength depending upon the amount added. Improved flexural strength is attributed to uniform distribution of the filler particles within the matrix and transformation toughening. Aluminium oxide exists in several crystalline phases, and all filler particles revert to the most stable hexagonal alpha phase at elevated temperatures. This is the phase of particular interest for structural applications [20,46]. When sufficient stress develops and micro-cracks begin to propagate, the transformation phenomenon occurs, which depletes energy for crack propagation [24].Therefore, proper distribution of the filler within the matrix can stop or deflect cracks [25]. Grant and Greener [46] previously demonstrated that even in the absence of coupling agent, incorporating sapphire whiskers (aluminium oxide) into heat cured denture base resin substantially increased the flexural strength compared with unmodified heat cured resin.

Reduced flexural strength at higher concentrations can be attributed to decrease in cross section of load bearing polymer matrix; stress concentration because of too many filler particles; changes in the modulus of elasticity of the resin and mode of crack propagation through the specimen due to an increased amount of fillers; void formation from entrapped air and moisture; incomplete wetting of the fillers by the resin; and the fact that aluminium oxide acts as an interfering factor in the integrity of the polymer matrix [17,18,19].

TABLE I
SUMMARY OF STUDIES FOR EFFECT OF ALUMINA ADDITION ON FLEXURAL STRENGTH

Authors (year)	Percentage of Alumina (wt%)	Effect on flexural strength
Jasim B S and Ismail I J (2014) [47]	1,2,3	Sig. inc. at 1%, dec. at 3%
Saritha M K et al(2012) [48]	5,10,15	Sig. inc. at 10,15%
Chaijareenont P et al(2012) [49]	10	Sig. inc.
Vojdani M et al(2012) [50]	0.5,1,2.5,5	Sig. inc. at 2.5%, dec. at 5%
Yadav NS & Elkawash H(2011) [51]	5	Sig. dec.
Arora N et a(2011) [52]	25	Sig. dec.
Ellakwa A E et al(2008) [20]	5,10,15,20	Inc. upto 15%

Sig. inc. – Significant increase , Sig. dec. - Significant decrease

Thermal diffusivity

The improvement in thermal diffusivity of the PMMA resin upon addition of alumina can be attributed to the formation of thermally conducting pathways within the polymer matrix.

TABLE II
SUMMARY OF STUDIES FOR EFFECT OF ALUMINA ADDITION ON THERMAL DIFFUSIVITY/THERMAL CONDUCTIVITY

Authors (year)	Percentage of Alumina (wt%)	Effect on thermal diffusivity / conductvity
Jasim B S and Ismail I J(2014) [47]	1,2,3	Sig. inc.
Atla J et al(2013) [53]	5,10,15,20	Sig. inc.
Arora N et a(2011) [52]	25	Sig. inc.
Abdulhamed A N & Mohammed A M (2010) [54]	5,7.5,10	Sig. inc.
Ellakwa A E et al(2008) [20]	5,10,15,20	Sig. inc.
Ebadin B and Parkan M A(2002) [55]	15,20	Sig. inc.
Messersmith P B et al(1998) [56]	9.35,15 vol%	Sig. inc.

Sig. inc. - Significant increase

This may due to overlapping of thermal conductive particles inside the polymer matrix to bridge the insulating effect of PMMA matrix. The increase in the amount of fillers make the particles approximate from each other and increase overlapping of thermal conductive particles that form pathway and permit transition of heat from one side of specimens to another side thus increasing thermal conductivity [5,20,40,47].

Hardness

Addition of alumina in variable concentrations lead to an increase in hardness of acrylic resins.This finding is in agreement with previous investigators, [22,28] who have concluded that reinforcing dental restorative resins and acrylic resin with ceramic particles can produce some improvement in the surface hardness.

TABLE III
SUMMARY OF STUDIES FOR EFFECT OF ALUMINA ADDITION ON HARDNESS

Authors (year)	Percentage of Alumina (wt%)	Effect on hardness
Jasim B S and Ismail IJ(2014) [47]	1,2,3	Sig. inc.
Vojdani M et al(2012) [50]	0.5,1,2.5,5	Sig. inc. at 2.5 and 5%
Abdulhamed A N & Mohammed A M (2010) [54]	5,7.5,1	Sig. inc.

Sig. inc. – Significant increase

This increase in hardness may have been due to inherent characteristics of the Al_2O_3 particles. Al_2O_3 possesses strong ionic interatomic bonding, giving rise to its desirable material characteristics, that is, hardness and strength.The most stable hexagonal alpha phase Al_2O_3 is the strongest and stiffest of the oxide ceramics. Therefore, it is expected that when Al_2O_3 particles disperse in a matrix, they increase its hardness and strength [20,46].

Surface Roughness

Addition of alumina in PMMA does not adversely affect surface roughness. It may be attributed to fine paricle size and well dispersion. The surface roughness of denture material is important, because it affects the oral health of tissues in direct contact with the dentures [31,32].The surface roughness threshold for acrylic resin is 0.2 microns, below which no significant decrease in bacterial colonization occurs [57]. The surface roughness of polished acrylic resin varies between 0.03 microns and 0.75 microns. However, an important factor in the clinical performance of a material is the way it responds to hygiene procedures [58]. In agreement with the study of Saad-Eldeen et al (2007) [26], the results of various studies have shown that incorporating Al_2O_3 at different concentrations did not adversely affect the roughness of the denture base resin [57].

TABLE IV
SUMMARY OF STUDIES FOR EFFECT OF ALUMINA ADDITION ON
SURFACE ROUGHNESS

Authors (year)	Percentage of alumina (wt%)	Effect on hardness
Jasim B S and Ismail IJ et al (2014) [47]	1,2,3	No Sig. inc.
Vojdani M et al (2012) [50]	0.5,1,2.5,5	No Sig. Inc.
Abdulhamed A N & Mohammed A M (2010)	5,7.5,10	No Sig. inc. till 7.5%

Sig. inc. – Significant increase

Water sorption

The absorption of water by acrylic resins is a phenomenon of considerable importance. Acrylic resins absorb water slowly over a period of time, primarily because of the polar properties of the resin molecules. High equilibrium uptake of water can soften an acrylic resin because the absorbed water can act as a plasticizer of acrylic and reduce the strength of the material [34]. The mechanisms which responsible for the water sorption was diffusion which was defined as the migration of one substance through a space or within a second substance in which water will penetrate acrylic resin mass and occupy a position between polymer chains [35]. From the results of various studies ,there was significant decrease in the

TABLE IV
SUMMARY OF STUDIES FOR EFFECT OF ALUMINA ADDITION ON
WATER SORPTION

Authors (year)	Percentage of alumina (wt%)	Effect on water sorption
Jasim B A and Iismail I J(2014) [47]	1,2,3	Sig. dec.
Asa r N V et al(2013) [59]	2 **vol%**	Sig. dec.
Arora N et a(2011) [52]	0.5,1,2.5,5	Sig. dec.
Abdulhamed A N & Mohammed A M (2010) [54]	5,7.5,10	Sig. dec.

Sig. dec. - Significant decrease

value of water sorption reported as the percentage of Al_2O_3 particles increased. The reasons why Al_2O_3 added groups exhibited significantly lower water sorption values than control group can be explained in several ways. During the polymerization process of acrylic resins, porosity or microvoids can occur among polymer chains. A high level of porosity or microvoids has been shown to facilitate fluid transport into and out of polymer by serving as sites for molecules to be sequestered and leading to enhanced solvent uptake and elution. Al_2O_3 particles used are insoluble in water and could reduce the overall volume of the absorbing polymer. Solubility represents the mass of the soluble materials from the polymers. The soluble materials present in denture base resins are initiators, plasticizers, free monomer and some pigmentation [4]. There were significant decrease in the values of water solubility with the increase in percentage of silanized Al_2O_3 particles,this decreases could be attributed to the fact that Al_2O_3 is insoluble in water so that the addition of Al_2O_3 to the mass of the specimens would act as additives and their presence will lead to reduction in the solubility of acrylic resin.

III. Critical Appraisal of Research on Alumina Reinforcement

- **Particle size**

Different particle size has been used by various researchers which leads to variable results.however, it has been observed that range of particle size is more effective than uniform size of particles. Vojdani M et al (2012) used 5 wt% of 3 µm untreated particles which led to 5.82% decrease in flexural strength [50] . Saritha M K et al (2012) [48] used 5wt% of 5-22µm untreated particles which increased in flexural strength by 13.99%.Also,nanofillers are more effective at lower concentrations as compared to micro fillers. Jasim BS and Ismail IJ(2014) used 1wt% treated nanofillers and reported 24% increase in flexural strength [47]. Chaijareenont P et al (2012) used 10 wt% treated particles 8-23 µm which led to 23.86% increase in flexural strength [49].

- **Particle shape**

Different particle shape have also been used by various workers. It has been found that elongated particles are more efficient in improving the properties of acrylic resin than spherical particles in accordance with studies done by Messer smith PB et al (1998) and Ben Hasan SA et al (2014) [56,60].

- **Use of coupling agent**

Use of particles treated with silane coupling agent leads to marked increase in properties of acrylic resin as compared to untreated particles. Chaijareenont P et al (2012) used 10 wt % 18-23µm particles which when silanized led to 23.86% and non-silanized 5.46% increase in flexural strength [49].

- **Type of Silane Coupling Agent**

Various researchers /authors such as Jasim BS and Ismail IJ (2014),Chaijareenont P et al (2012) have recommended the use of the silane coupling agent

MPS (3- methacryloxy propyltrimethoxysilane) [47,61].

IV. GUIDELINES FOR FUTURE RESEARCH

- Studies based on shape and size of particles need to be conducted.
- Research based on type and amount of silane coupling agents need to be conducted.
- Elaborate studies to evaluate the effect on other properties of acrylic resin need to be conducted.
- Long term Clinical studies and studies based on patient satisfaction need to be conducted.

V. CONCLUSION

Present literature highlights the positive impact of alumina addition on the physical and mechanical properties of the acrylic resin. However, further laboratory and clinical research is required before this reinforced acrylic can be successfully used as a denture base material.

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