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Conventional and Contemporary polymers for the fabrication of denture prosthesis: part I – Overview, composition and properties

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Abstract
Polymers are widely used in dentistry for various applications. Dentures and dental implants are the major prosthetic devices given to restore physiological and esthetic functions of oral tissues of edentulous or partially edentulous patients. Complete and partial dentures based on polymeric compositions are the most popular devices since the cost of dental implants and metal-base dentures are much higher. This article reviews polymeric denture base materials with respect to their composition, chemistry, properties along with their pros and cons from the literature available in PUBMED and other available sources from the past 50 years.

Keywords: Dentures, Vulcanite, Bakelite, Celluloid, PVC, PMMA.

Introduction
Tremendous developments in the science of biomaterials over the last few decades largely contributed to the increase in the life expectancy of human beings. Dental materials are classified into four major groups such as metals, ceramics, polymers and composites. Polymeric based compositions are widely used for the fabrications of complete and partial dentures. In addition, denture soft liners, resin cements, pit and fissures sealants also consist of polymers. Although, dental implants have received large attention with a high success rate for the treatment of complete and partially edentulous conditions, dentures remain the most popular choice of prosthetic devices [1, 2, 3]. Complete dentures are conventionally constructed with some polymers [1, 2, 4, 5], precious metal alloys [1, 2, 6, 7] and base metal alloys [1, 2, 4, 8-11].
History of complete dentures for the treatment of edentulism dates back to 700 BC. Various materials such as bone, wood, ivory, and vulcanized rubbers were utilized to fabricate complete dentures. Figure 1 describes the development of polymers used for the fabrication of denture prosthesis. During the early 1900's, materials such as poly vinyl chloride, vinyl acetate, modifications of bakelite and cellulose plastics were used [5, 12]. In 1937, "*Walter Wright*" introduced Poly (methyl methacrylate) [PMMA] material as denture base material and it has become the most commonly used to fabricate complete dentures and removable partial dentures [13, 14]. Ideally, a denture base material should be biocompatible with the oral tissues and also should be able to with stand the masticatory forces [1, 2, 4, 14]. The ideal characteristics of a denture base material are detailed in table 1 [1-4, 15, 16]. The main objective of this article is to review the composition, chemistry, and properties of polymeric based materials used for the fabrication of denture bases.

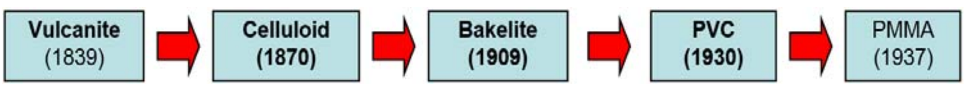

Fig 1: Development of polymerns for the fabrication of denture bases

2. Vulcanite
Vulcanite was one of the first polymers to be used successfully as a denture base material. It was introduced by Charles Goodyear in 1839. The invention of vulcanite distinguished a

considerable increase in demand for accurately fitting prosthesis (Figure 2) at reasonable cost [17, 18].

Fig 2: Complete dentures made with Vulcanite polymer

Vulcanite is formed by the addition reaction of natural rubber and sulphur. The production of vulcanite was carried out in a vulcaniser under steam pressure at 160 to 170 °C [19]. The amount of sulphur modifies the hardness of vulcanite. Sulphur helps in cross linking between the rubber polymer chains (figure 3) to form a rigid, opaque and stable solid [5, 19]. Vulcanised rubbers exhibit superior hardness, elasticity, heat resistance, high melting point and more resistance to oxidation than the natural rubbers. However, they lack adequate translucency that results in poor aesthetics and also the porcelain teeth needs to be mechanically retained to it due to the lack of bonding. Mechanical retention can be achieved by making undercut holes in the posterior teeth and pins in the anterior teeth [20]. Vulcanite was porous leading to the accumulation of plaque and oral fluids resulting in an unhygienic denture base [18, 21, 22].

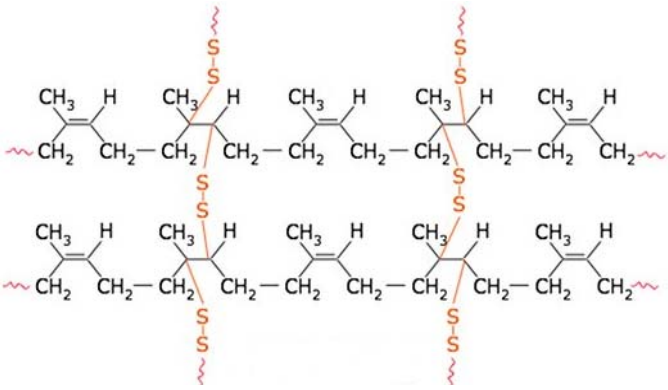

Fig 3: Vulcanized rubber polymer with di-sulphide cross-links.

3. Celluloid

Celluloid is a natural cellulose polymer and it was introduced in circa, in 1870. Celluloid can be produced by plasticising cellulose nitrate with camphor after which the pigmentation is carried out. A denture base is constructed by pressing the celluloid blank into a dry, heated mould [5]. Celluloid was considered to be a promising alternative to the widely used vulcanite. However, popularity of celluloid was soon diminished as it was found to rapidly discolour over time, absorbing water and stains from food, drinks and tobacco [5, 18], persistent residual camphor taste from the denture base. Further, it was difficult to repair a fractured denture [17, 23, 24].

4. Phenol-formaldehyde (Bakelite)

Phenol formaldehyde resin was discovered by the Belgian chemist Dr. Leo Baekeland in 1909. Phenol formaldehyde resin was also called as 'Bakelite' and first produced for commercial use in dentistry in 1924 [5, 17, 18, 25]. Bakelite is made by condensation polymerization which involves condensing one or more types of phenols with formaldehyde, as shown in figure 4. Bakelite denture bases were having excellent aesthetics immediately after processing. However, persistent taste of phenol and staining very early are the drawbacks of this material. Furthermore, phenol-formaldehyde denture bases were very brittle and prone to fracture [17]. They also are very difficult to repair and exhibited poor shelf life [24, 25].

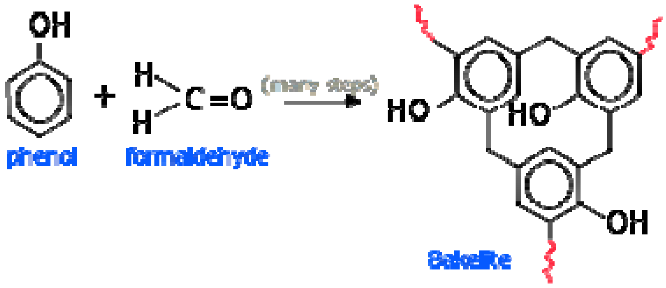

Fig 4: Several units of phenol and formaldehyde combine to produce bakelite polymer

5. Polyvinyl chloride (PVC)

In 1930s, a co-polymer of vinyl chloride (80%) and vinyl acetate (20%) was introduced as a denture base material. The structure of PVC is shown in figure 5. The processing method to fabricate the denture was similar to celluloid [18, 24]. The residual stresses may be introduced during processing that result in gradual deformation of the denture base leading fracture during service [24]. In addition, PVC may also discolour at temperature used during fabrication.

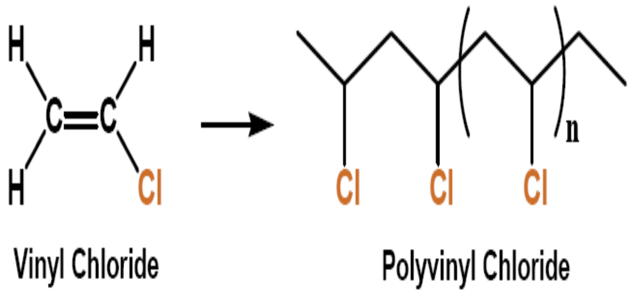

Fig 5: Structure of Polyvinyl Chloride

However, PVC is still used currently as a denture lining material and also for construction of athletic mouth guards. PVC was plasticized with either dibutyl or dioctyl phthalate for this purpose. PVC is available as pre-plasticized sheet, which can be used to construct protective mouth guards. Processing generally involves heating the Pre-plasticised sheet followed by moulding it to the desired contour with the use of a vacuum to seal the sheet of material over a cast of the patient's teeth [26]. Even though the material is still used for this purpose, the material's properties are far from ideal. They harden over time as the plasticiser leaches out during service [2, 27]. In addition, they are difficult to polish. This results in poor denture hygiene and acts as an irritant to the oral mucosal tissues [22, 28-30].

6. Poly (methyl Methacrylate)

No denture base material was able to fulfill the ideal requirements discussed in table 1. However, it was in 1937, "*Walter Wright*" introduced Poly (methyl methacrylate) [PMMA] material as denture base material and it was found to be the most superior material over all other denture base materials [14]. It became so popular that during 1940s' almost all the dentures were fabricated with acrylic based materials [14, 18, 31]. Although, several new materials such as polystyrene and light-activated urethane dimethacrylate were introduced, PMMA remained the most preferred material of choice for both complete and partial denture prostheses [1-4, 14, 32]. PMMA is the polymer of methyl methacrylate, with chemical formula $(C_5H_8O_2)_n$. It is a clear and colourless polymer.

Table 1: Ideal requirements of Denture Base Materials

Property	Requirements
Biological	• Should be non-toxic, non-irritant and non-carcinogenic
Chemical	• Should be insoluble in the oral fluids or any other fluids being taken the patient. • Should not absorb oral fluids or any other fluids being taken by the patient as it causes dimensional changes. • Should adhere very well with artificial teeth and liners.
Mechanical	• Modulus of elasticity should be high. It enables the denture base to be rigid against masticatory forces. • Resilience should be high in order to protect the underlying soft tissues by absorbing masticatory forces. • Should have high elastic limit and proportional limit to prevent the permanent deformation when stressed. • Should have adequate mechanical strength to resist fracture under repeated masticatory forces. • Should be dimensionally stable. • Should have adequate abrasion resistance. • Specific gravity should be low (especially for maxillary dentures).
Thermal	• Should be a good thermal conductor. • Co-efficient of thermal expansion (COTE) should match with that of the artificial teeth. • Softening temperature should be more than the boiling temperature of the water.
Esthetic	• Should exhibit sufficient translucency so that it can be made to match the appearance of the oral tissues. • Should be capable of tinted or pigmented.
Other	• Should maintain the desirable properties for extended period of time after manufacture. • Should be inexpensive. • Should be easy to manipulate. • Should be radio-opaque so that they can be detected if some part of the denture is accidentally swallowed. • Should be easy to repair. • Should be easy to clean. • Should have longer shelf life.

PMMA is routinely produced by suspension or emulsion polymerization [33]. PMMA used in dentistry is produced by suspension polymerization. Generally, PMMA is formed by addition polymerisation of multiple methylmethacrylate monomers in the presence of an initiator, typically benzyl peroxide (BP) [2, 34]. In the presence of heat or chemical activation, BP breaks down to produce free radicals. These free radicals act upon the vinyl group of methyl methacrylate, opening the double bond leading to the formation of a new single carbon-carbon bond (figure 6) [1, 2, 35, 36]. The free radical formed from the methyl methacrylate double bond is asymmetrical. This results in a carbon atom that also has an asymmetrical environment after reaction. The resulting polymer is atactic [37]. There are three types of PMMA denture base materials based on their activation of polymerization mechanism they include heat activated, chemically activated and light activated PMMA denture base resins [2].

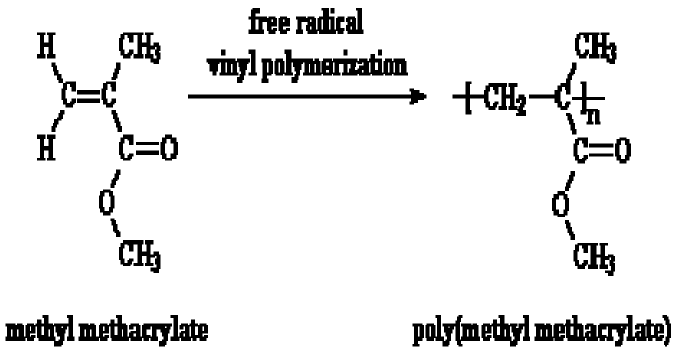

Fig 6: Preparation of PMMA from MMA using free radical vinyl polymerization.

6.1. Heat activated PMMA Resins

These materials are widely used for the construction of removable complete or partial dentures. In heat-cured materials, the polymerization process is initiated by the release of free radicals from BP on supply of heat energy [2, 38]. These free radicals react with the monomer molecules and continue till the monomer is available [2, 39]. These materials are supplied in the form of powder and liquid. Powder contains finely divided pre-polymerized PMMA beads and liquid contains methyl methacrylate (MMA). Liquid is supplied in a dark brown colored bottle to avoid accidental polymerization when exposed to visible or U.V. radiation during transport and storage. Bottle should be kept closed to prevent evaporation [2]. The detailed composition is presented in table 2.

Table 2: Composition of Heat activated PMMA [1, 2, 8, 35]

Powder	Liquid
• Pre-polymerized poly (methyl methacrylate). • Copolymers of of PMMA (5%) E.g.: Ethyl or Butyl methacrylates • Initiator (0.2-1.5%) E.g.: benzoyl peroxide. • Plasticizer E.g.: Dibutyl phthalate. • Color pigments E.g.: Mrcuric sulphide, Cadmium Sulfide, Ferric oxide, Carbon Black. • Opacifiers E.g.: Zinc or titanium oxides • Dyed synthetic fibers made from nylon or acrylic • Inorganic particles E.g.: Glass fibers, zirconium silicate, whiskers of alumina, SiC, boron nitride and carbon fibers. • Heavy metal compounds E.g.: Barium, bismuth, etc.	• Methyl methacrylate • Co-monomers • Inhibitor E.g.:Hydroquinone (0.003%-0.1%). • Plasticizers E.g.: Butyl or Octyl methacrylate and Dibutyl Phthalate. • Cross-linking agent E.g.: Ethylene glycol dimethacrylate

6.1.1. Properties of methyl methacrylate

Methyl methacrylate is a clear, transparent liquid at room temperature. Monomer is a volatile liquid with distinct odour exaggerated by a relatively high vapour pressure at room temperature. The detailed properties of MMA are discussed in table 3.

Table 3: Properties of MMA

Properties of MMA	
Melting point	-48 ^{0}C
Boiling point	100.8 ^{0}C
Density	0.945 g/cc at 20 ^{0}C
Polymerization shrinkage	21%
Heat of polymerization	12,900 cal/mole

6.1.2. Properties of Poly (methyl methacrylate)

Completely polymerized Poly (methyl methacrylate) is highly transparent just like glass. Most important concerns for polymers are the monomer to polymer conversion and residual monomer content because of their application in approximation to oral tissues. Acrylic resin dentures contain methyl methacrylate as residual monomer which has the potential to elicit irritation, inflammation and allergic response of the oral mucosa [18, 40-43]. Further, residual monomer is capable of producing both stomatitis and an angular cheilitis [40, 42, 44-52]. Residual monomer present in heat cure acrylic resin is 0.2 – 0.5% and in self-cure acrylic resin, it is 2 – 5% [2, 53]. The effects of residual monomer leached on tissues have been reported by clinical observations [54], in animal models [55], and by in vitro cell growth [56–58]. Lyapina *et al*., 2014, studied and evaluated the incidence and performed a comparative risk assessment of cross-sensitization to methacrylic monomers in different groups of individuals. This study clearly indicated that the high incidence of sensitization to methacrylic monomers in dental practice, with cross-sensitization was clearly manifested [43].

Formaldehyde is another allergic agent in acrylic dentures responsible for mucosal injuries. Formaldehyde is formed as an oxidation product of the residual MMA monomer in inhibition layers and poorly polymerized resins. Formaldehyde formation was suggested to occur through the decomposition of the oxygen-methyl methacrylate copolymer or by the oxidation of methyl methacrylate. Formaldehyde is proved to be cytotoxic even at much lower concentrations than methyl methacrylate. Formaldehyde is also a strong irritant to the mucous membranes even at concentrations as low as 0.63 to 1.25 mg/m^3 [38, 59-63].

The allergic reaction occurs with in a few to several hours after the mucosa is exposed to the resin. When allergic reactions were noted, they were described as white, necrotic lesions on the mucosa; either as small, multiple lesions or as large ulcers mimicking allergic stomatitis [38, 42, 61-65]. Toxic and allergic effects of acrylics depend on several variables including polymer: monomer ratio, storage time and water immersion, type of polymerization cycle and polymerization method used [41]. The lesser the polymer: monomer ratio, the greater the amount of residual monomer and, therefore more potential for cytotoxicity [66]. The cytotoxic effect of acrylic resins was greater in the first 24 hours after polymerization and decreased with time. Therefore, the longer a denture is soaked, the less cytotoxic effect irrespective of type of the denture base resin used [44]. Longer the polymerization cycle lesser is the residual monomer. Therefore less cytotoxic effects can be observed [2].

Acrylic resins are soluble in aromatic hydrocarbons, ketones and esters and quite less soluble in water comparatively. Alcohol functions as a plasticizer, and reduces the glass transition temperature. Therefore, solutions containing alcohol should not be used for cleaning or storing the dentures. The sorption of water by the PMMA changes the dimension of the denture when alternatively soaked in water and dried which leads to crazing [1, 2, 67, 68]. Chemical and mechanical properties are detailed in table 4.

Table 4: Properties of heat activated PMMA resin

Solubility	**In aromatic hydrocarbons, ketones and esters.**	**0.04 mg/cm^2**
	In water	**0.02 mg/cm^2**
Water sorption		0.69 mg/cm^2
Modulus of elasticity		3.8X10^3 Mpa.
Proportional limit		26 Mpa.
Compressive strength		76 Mpa
Tensile strength		48 – 62 Mpa
Percentage of elongation		1-2%
Impact strength		0.98 – 1.27 J
Surface hardness		18-20 KHN
Density		1.16 – 1.18 g/cc
Fatigue strength		1.5X10^6 cycles at 17.2 MPa
Thermal conductivity		5.7X10^{-4} ^{0}C/Cm
Coefficient of thermal expansion		81X10^{-6} / ^{0}C
Heat distortion temperature		71 – 91 / ^{0}C
Glass transition temperature		125 ^{0}C
Depolymerization temperature		450 ^{0}C

Ideally, a denture's surface should be free of roughness. The rough surfaces of denture may damage the adjacent soft tissues and also encourage the growth of the bacteria [53, 69]. It has been discussed in the literature that the clinically acceptable threshold level of surface roughness (Ra) is 0.2 µm [53, 70]. Hardness provides a possible indication of the abrasiveness of the denture material [53]. Conventional dentifrices and cleansing agents are contra-indicated for cleaning PMMA dentures as it has very less hardness.

The popularity of PMMA as denture base material was attributed to its ease of processing, low cost, lightweight, excellent aesthetic properties [10, 14, 71, 72], low water sorption and solubility; and ability to be repaired easily. However, low thermal conductivity, inferior mechanical strength, brittleness, high coefficient of thermal expansion and relatively low modulus of elasticity makes it more prone to failure during the clinical service [10, 12, 14, 73, 74].

Clinical failure of PMMA dentures are most likely in the form of fracture either due to fatigue [75] or impact forces [76]. Flexural fatigue of dentures as evidenced by midline fracture is due to the stress concentration around the micro cracks formed in the material due to continuous applications of small forces. Repetitive nature of masticatory load results in propagation of cracks which weakens the denture base and finally results in fracture [14, 77, 78]. Fracture of dentures by impact forces, on the other hand, results from the sudden application of force to the dentures. Such types of fractures are more likely due to the accidental dropping of dentures on surfaces during cleaning of dentures by patients [2, 12, 14, 79, 80]. Fracture of dentures in clinical service has been a concern and several attempts have been made to improve flexural and impact strength of PMMA. Research in this area aimed at modifying the composition or reinforcing the PMMA with stronger materials and developing new materials with better properties [14]. The approaches include modifying the composition with copolymers [2, 14], reinforcing the dentures with various fibers such as aramid fibers [14, 81], carbon / graphite fibers [14, 82, 83], polyethylene fibers [13, 14, 84, 85], glass fibers [14, 86-94] and also reinforcing with various metallic fillers [14, 81, 95, 96] and nanoparticles such as silica and titania nanoparticles [14, 86, 97-99]. The modifications and recent advances in PMMA based denture base resins will be discussed in part II.

6.2. Chemically activated PMMA denture base resins

The resin materials which polymerize without the application of heat or light are called chemically cured resins, i.e.; polymerization reaction is initiated by a chemical activator (N, N – Di Methyl P- Toludene) and the polymerization reaction completes at room temperature. These resins were first used for dental purposes in Germany during world war-II and are known variously as self-cure or cold cure or auto polymerizing resins. These are supplied as powder and liquid form [2]. The typical composition of chemically activated resin is given in table 5.

Table 5: Composition of chemically activated PMMA

Powder	*Liquid*
Poly (Methyl Methacrylate) or Copolymer beads. Benzoyl peroxide (maximum – 2.0%) – Initiator. Pigments Colored fibers (Nylon/acrylic) – added for aesthetic effect.	Methyl Methacrylate Monomer. Ethyl Glycol Di Methacrylate – cross linking agent. Hydroquinone – inhibitor. Dibutyl Phthalate. Tertiary amine (Di Methyl Para Toludene) – Activator (maximum – 0.75%)

6.2.1. Properties

Unlike, heat curing resins, the polymerization of self-curing resins is never complete. Self cured resins are composed of 3 to 5% of residual monomer [2, 53]. Free monomer may be released from the denture and irritate the oral tissues (potential irritant). Residual monomer will act as a plasticizer and makes the resin weaker and more flexible (decreases transverse strength). Chemically activated resins display slightly less shrinkage than their heat activated counterparts, because of a less complete polymerization. This imparts greater dimensional accuracy to chemically activated resins [2].

The color stability of chemically activated resins generally is inferior compared to heat-activated resins. Tertiary amine based activator present in the resin is susceptible to oxidation and accompanying color changes that may affect the appearance of the resin. Discoloration of these resins may be minimized by the addition of stabilizing agents to prevent such oxidation. Activators like organic acids produce products with improved color stability, but the compounds are not chemically stable [1, 2, 22, 23].

7. Other Polymer materials

Various polymer based denture base materials such as polypropylene, polystyrene, epoxy resin, polycarbonate, and polyethers have been developed as alternatives to PMMA resins. However, none of these alternative materials could be used as absolute substitutes of PMMA. Epoxy resin denture base has higher water sorption, low fracture strength, heavy deposition of stains and calculus [17].

8. Conclusion

PMMA, though not ideal, is still considered to be the material of choice among various polymers. The flexural and impact strength of PMMA resins have been improved by chemical modification of existing PMMA and also by reinforcing with various fillers such as metallic powders, fibers, nanoparticles, and nanorods. The modifications in the PMMA denture base resins will be discussed in Part II.

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