

Factors Affecting Sorption and Solubility of Denture Base Acrylic Materials: A Review

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KEYWORDS

Surface treatment, polymethyl methacrylate, polymerisation, heat-cure, artificial saliva

ABSTRACT

Heat-cured PMMA is one of the most frequently used materials in the manufacture of removable dentures. Heat-cured material used to fabricate dentures contain soluble elements, allowing the material to absorb and release substances. These factors all directly affect the longevity and performance of prostheses fabricated from polymethyl methacrylate denture base material. Various procedures associated with fabrication of removable dentures from heat-cure denture base material are reported to significantly influence the sorption and solubility experienced by the prosthesis. A comprehensive search of peer-reviewed journals located within academic databases was conducted to identify relevant literature pertaining to sorption and solubility of denture base materials. Key aspects of each paper were captured in Microsoft® Excel® to record author/s, location, study design, sample size, methodology, results and conclusions. Mixing ratios and polymerisation cycles have been identified as variables that can negatively influence the rate of sorption and solubility of denture base materials during the fabrication process. Certain surface treatment procedures, as well as storing the denture in artificial saliva solution may reduce the levels of sorption and solubility experienced and optimise its lifespan. In this review we identify these factors and consider the detrimental effects of sorption and solubility on denture base acrylic materials.

INTRODUCTION

Heat-cured polymethyl methacrylate (PMMA) remains the denture base material of choice for many due to its excellent aesthetics, ease of processing and repair, and relative affordability [1]. Although PMMA exhibits several properties of an ideal denture base material, drawbacks such as residual monomer toxicity [2], a high coefficient of thermal expansion, poor mechanical strength as well as dimensional instability [1] have drawn criticism. One of the main contributing factors to PMMA acrylic's limitations is the resin's ability to absorb water when subjected to a moist environment, which causes dimensional instability

[3]. Furthermore, the sorption and solubility of denture base materials do not only affect the material's mechanical properties as the release of unreacted monomer and other water-soluble by-products from the denture base material during function may also result in an allergic reaction of the oral mucosa [4]. Because of this, the water sorption and solubility of denture base materials should be as low as possible. Mixing ratios, polymerisation cycles, the medium in which the material is submersed, as well as surface treatments have been identified as factors that are controlled by the user of the material that can influence the sorption and solubility of the denture base material. Since sorption and solubility both critically affect the clinical and mechanical performance of denture base materials, it was decided to summarise the available literature pertaining to these phenomena and provide a concise review.

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MATERIALS AND METHODS

This review was established through a comprehensive search of literature pertaining to the sorption and solubility of denture base materials from September 1976 to August 2020. A search of journal articles located within academic databases was conducted using the following keywords: Surface treatment, polymethyl methacrylate, polymerisation, polishing, heat-cure, mixing ratio and artificial saliva. The following databases were used: Google Scholar, Medline, PubMed, Wiley Online Library, Science Direct. Key aspects of each paper were captured in Microsoft® Excel® to identify the aim of the study and to record authors, location, study design, sample size, methodology, results, and conclusions. A total of 57 articles were identified of which 31 were included for the purposes of this review. Literature from peer-reviewed journals published within the last decade was primarily sought, but older studies as well as those listed on University databases were accepted if they met the inclusion variables relevant to the topic. As articles specifically relating the variables investigated in this review are not in abundance, a selection criterion that is of a too stringent nature could not be applied. The keywords were searched as individual factors and the articles were selected on the following criteria: relevance to heat-cure denture base acrylic, date of publication, English language and articles presenting comparative evaluations between different factors relating to this review.

SORPTION OF DENTURE BASE MATERIALS

When reviewing denture base materials, sorption is a crucial property to consider. High sorption values may have detrimental effects on the mechanical properties of a denture as well as reduce its longevity. Sakaguchi & Powers [5] explain that sorption is a process of adsorption and absorption. Adsorption is the molecular adhesion of a substance to the surface of a material, whereas absorption is the diffusion of a substance into the body of the material. When both are taking place, the term sorption is used to classify the phenomenon. Sorption occurs when a denture base is exposed to a moist environment and is initiated by the polarity of the PMMA's molecules. PMMA materials contain carbonyl groups, to which water molecules have an affinity [6]. Water molecules are adsorbed to the surface of the material and are further absorbed into the body of the denture base through porosity and intermolecular spaces via diffusion [3]. According to Ferracane [7], the research conducted by Braden et al. [8], Kalachandra & Turner [9] and Sideridou et al. [10] Ann Dent UM. 2022, 29: 1-8

all indicate that water sorption follows Fick's law of diffusion, which is based on the movement of molecules from a high concentration to a low concentration. The extent and rate of sorption experienced by the material are influenced by the density of the polymer network and the potential for hydrogen bonding and polar interactions to take place [7]. As water diffuses into the molecular structure of the PMMA material, its molecules occupy the intermolecular voids between the polymer chains [3,6]. Due to the weak secondary bonds in PMMA materials, the ingress of water molecules can force the polymer chains apart, resulting in the expansion of the polymerised material [11]. Polymers with a high cross-linking density have illustrated lower sorption values due to the limited free space available to be occupied by water molecules within the structure of the network [7]. The sorption of water into the structure of PMMA is an alternating event. When the material is exposed to an open and dry environment, it allows water to leave its structure and the material undergoes contraction [12]. This ongoing dimensional change and the instability caused by continuous expansion and contraction creates internal stresses within the material, which may result in the crazing and fracture of the denture [4]. Surface cracks may develop as a result of dimensional instability that can form points of entry or attachment for various bacteria, yeasts and moulds [13]. The interaction of water molecules with the polymer chains of PMMA may also result in the plasticisation of the denture base, affecting the mechanical properties of the material by decreasing its hardness, fatigue limit and transverse strength [4,14]. According to ISO standards 20795-1:2013(E), the water sorption of heat-cured acrylic should not exceed 32 µg/mm³ [20].

SOLUBILITY OF BASE MATERIAL BY-PRODUCTS

The solubility of denture base materials directly impacts the biocompatibility of the prosthesis, as high levels of diffusion may increase the risk of various cytotoxic effects. The importance of reducing the levels of solubility in a prosthesis cannot be stressed enough. According to Helmenstine [15], "solubility is defined as the maximum quantity of a substance that can be dissolved in another". PMMA denture base material should be insoluble with a molecular network of high chemical and thermal stability [7]. Anusavice et al [3] claim that PMMA denture base resins are insoluble in water, as well as in most substances that may be found in the oral cavity. Both Tuna et al [4] and Ferracane [7] elaborates on this and explain that even though PMMA denture base materials are insoluble in the oral

environment, they contain by-products such as unreacted monomer, plasticisers and other water-soluble additives that can absorb water and chemicals from the environment and release them into the oral cavity. The literature suggests that unreacted residual monomer is one of the primary soluble substances released from PMMA denture base material during function [16]. The solubility of such materials needs to be as low as possible to prevent the patient from experiencing cytotoxic effects such as erythema, oedema and urticaria [17]. Both the uptake and release of substances from denture base material occur as a result of diffusion. Klazema [18] describes diffusion as a process where substances such as water molecules and ions enter and leave the molecular structure of materials. This movement is enabled by a concentration gradient, resulting in molecules moving from an area of high concentration to an area of low concentration. This process is said to continue until the solute is evenly dispersed between the two materials. The process is also governed by the diffusion coefficient of the medium. The diffusion coefficient is a value indicating the rate at which diffusion can take place and is influenced by the temperature and viscosity of the medium [19]. For denture base polymers, the loss in mass (soluble matter) should not exceed $1.6 \mu\text{g}/\text{mm}^3$ for types 1, 3 and 5 polymers and should not exceed $8.0 \mu\text{g}/\text{mm}^3$ for type 2 polymers [20].

MIXING RATIOS

Manufacturers of PMMA denture base material all recommend different powder/liquid mixing ratios for their respective products to achieve optimum results. However, the modification of these mixing ratios by dental laboratory technicians attempting to manipulate the handling properties of the material is not uncommon. Additional monomer can be added to the mixture to extend the working time of the material, but a higher monomer concentration can lead to a greater amount of residual monomer present in the material after polymerisation. The addition of extra monomer is of concern as it may increase the chances of patients experiencing various cytotoxic effects from the denture base material during use [21]. Tuna et al [4] investigated the sorption and solubility of various acrylic resins and their results indicated that heat-cured acrylic resins had lower solubility values than most of the self-cured acrylic resins. In a similar study, Kostić et al [22] investigated the residual monomer content present in dental acrylic polymers and its effect on tissue after implantation. The materials used in that study were Triplex Cold and – Hot from Ivoclar Vivadent™, with a liquid to powder ratio of 10ml:13g and 10ml:23.4g Ann Dent UM. 2022, 29: 1-8

respectively. The authors recorded similar results to those of Tuna et al [4]. The residual monomer content present in cold polymerised PMMA was higher than that of heat polymerised PMMA, with cold polymerised material also showing a greater inflammatory response of soft tissue. Tuna et al [4] noted that as the primary difference between heat- and self-cured materials is the liquid-to-powder ratio, and taking the study by Kostić et al [22] into account, it can be confirmed that the amount of unreacted monomer present in the material after polymerisation directly affects the degree of solubility expressed by the material. These observations may not only be as a result of different liquid-to-powder ratios, as both Tuna et al [4] and Kostić et al [22] compared the heat-cure acrylic to self-cure acrylic. Heat-cure and self-cure acrylics do not only have different liquid-to-powder ratios, but different activators and polymerisation procedures as well. Nisar et al [23] investigated the effect of varying powder liquid ratios and curing conditions on the residual monomer concentration of heat cure acrylic resin. Their observations were in line with those of Jerolimov [24] who concluded that the polymerisation cycle has a greater influence on the residual monomer content of the material, than its mixing ratio. It is evident that the mixing ratio of the material and the polymerisation cycle operate in tandem and have a symbiotic effect on the residual monomer content post-processing. Kostić et al [22] explains that the residual monomer present in the material after polymerisation affects its molecular weight and density, resulting in a more porous structure. The excess monomer will leach from the material into its surroundings, resulting in intermolecular voids. The leaching of the monomer will be compensated by the uptake of water, leading to the plasticisation of the material. It can therefore be advisable to always follow the manufacturer's recommendations strictly regarding the use of the material to ensure optimum results.

POLYMERISATION CYCLE

Anusavice et al [3] characterise polymerisation as “a chemical reaction in which monomers of a low molecular weight are converted into chains of polymers with a high molecular weight”. The polymerisation of denture base polymers is induced by an initiator which is part of the material's chemical composition. Initiators react when exposed to external trigger mechanisms such as light or heat, dependent on the material. Various studies mention that incorrect polymerisation cycles can negatively impact the physical, mechanical, and biological properties of the denture. Each manufacturer recommends a

polymerisation cycle that is time and temperature orientated for a specific material.

Heat-activated PMMA denture base material is polymerised by addition polymerisation [11]. Ouellette & Rawn [25] explain that the process of addition polymerisation involves a chain reaction during which one carbon-carbon double bond adds to another. When using heat-activated PMMA, the mould containing the polymer–monomer mixture is heated in a water bath to activate the benzoyl peroxide initiator. The benzoyl peroxide breaks down to produce free radicals [23]. The free radical with its unpaired electron acts on the vinyl group of the methyl methacrylate molecule. The double bond of the methyl methacrylate molecule is split, resulting in the free radical forming a single bond with the monomer on one side, while the remaining free electron remains unpaired. This results in a radicalised monomer molecule [3,11,26,27]. The radicalised monomer interacts with other methyl methacrylate molecules in the network. Its unpaired electron interacts with the double bond of the remaining methyl methacrylate molecules, initiating a chain reaction where the remaining monomer molecules attach to a radicalised monomer chain. This occurs at various sites in the network and results in the formation of many radicalised monomer chains. This process is known as chain growth [3,11,26]. Additional polymerisation does take place not only between molecules, but between radical chains as well. Covalent bonds form between the integrating radicalised monomer chains resulting in the formation of large macromolecules. This process continues until the monomer molecules in the network have been exhausted [2,3]. As the polymerisation process continues, the concentration of free radicals and monomer molecules decrease and the structure of the mixture becomes more viscous, causing the rate of polymerisation to slow down. To ensure maximum polymerisation, the temperature of the polymerisation cycle is usually increased as time progresses as it supplies more energy to the system, allowing the molecules to move more rapidly, and in turn increases the degree and rate of polymerisation. However, not all the monomer molecules are polymerised during the polymerisation process and results in unreacted residual monomer being present in the polymerised material. The unreacted monomer may leach from the denture base during function, resulting in cytotoxic effects in the oral cavity [2].

Altering the polymerisation cycle may lead to insufficient monomer-polymer conversion or overheating of the monomer, resulting in an

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increase in residual monomer present after polymerisation or porosity [6]. Examination of the literature indicates that both residual monomer and porosity may increase the sorption and solubility experienced by denture base acrylics. Tuna et al [4] reported that materials with a homogenous structure are less susceptible to sorption and solubility and that high porosity values would enhance this phenomenon. The prementioned literature is supported by the findings of Wang et al [28], Bayraktar et al [29] and Nisar et al [23] who all confirmed that variations in the time and temperature ratio of polymerisation procedures effect the sorption and solubility of the material post-processing. In practice, many laboratory technicians alter the processing procedure to save time or money. These actions may have detrimental effects on the physical, mechanical, and biological properties of the material. It is therefore advised to always adhere to the manufacturer's recommendations, which have been precisely calculated to ensure optimum results.

SURFACE TREATMENTS

When dealing with denture base polymers, surface treatments are applied to improve the properties and characteristics of the material, whether for physical, mechanical, chemical or aesthetic purposes. PMMA denture base surfaces that are highly polished and smooth promote increased gingival health, including chewing efficiency, patient comfort, improved aesthetics and prosthesis longevity. Mechanical polishing is the conventional polishing technique, making use of abrasives of varying degrees to alter the surface of the material by reducing its roughness or texture [30]. Chemical polishing is an alternative method which reduces the overall polishing time and is able to reach areas of the denture that are not accessible to mechanical polishing. When making use of chemical polishing, the prosthesis is placed in a chemical polisher containing heated methyl-methacrylate, after the finishing procedures [31,32]. Rahal et al [31] explain that the molecules from the heated methyl methacrylate polishing liquid penetrate the superficial polymeric chains of the acrylic, breaking the secondary bonds that join them. This results in a final plasticizing effect of the acrylic resin.

Various studies have investigated the efficiency of mechanical and chemical polishing techniques and their effect on the surface roughness of denture base acrylic resins. Authors such as Rahal et al [31], Al-Kheraif [33] and Al-Rifa'i [34] all found that mechanical polishing is more effective in reducing

the surface roughness of denture base materials than the chemical alternative. They therefore concluded that mechanical polishing is a more effective polishing technique. A polishing procedure may not only help reduce the surface roughness of denture base polymers but may also have a significant effect on the sorption and solubility levels of the material. A study conducted by Engelbrecht [35] investigated the factors influencing the sorption, solubility and cytotoxicity of a heat-cured denture base polymer. The results indicated that the conventional polishing procedure reduced both the sorption and solubility values of the material, but that this reduction was only statistically significant for solubility. The author speculated that these occurrences could be attributed to the different chemical processes and the polarity and size of different molecules involved in the processes of sorption and solubility. These results were in agreement with Al-Muthaffar [36], who aimed to determine the effect of a conventional polishing procedure on the water sorption of cold- and heat-cured acrylic denture base material. Al-Muthaffar [36] found that the conventional polishing procedure significantly reduced the amount of sorption experienced by both cold- and heat-cured acrylics. He reasoned that the observed results might have occurred because the heat generated during the conventional polishing procedure often exceeds the glass transition temperature of the acrylic, which could result in the smearing of the resin's surface. The smeared surface decreases the polarity of the acrylic by minimising the concentration of polar sites available to form hydrogen bonds with water molecules. The irregular surface of the unpolished specimens means that their surface area is greater than that of the polished specimens, resulting in a greater interface area between the specimens and water molecules. This greater contact may lead to greater water uptake. It can also be explained in terms of the contact angle hysteresis between the water droplets and the surfaces of the samples. Rahal et al [37] referenced Monsé né go et al [38], who explained that water droplets form lower contact angles with rougher surfaces. Surfaces that produce lower contact angles are of a more hydrophilic nature and therefore increase the material's affinity to water.

Mechanical and chemical polishing procedures are not the only surface treatments that may improve the characteristics and properties of denture base materials. The application of light-cured varnish products to the surface of PMMA denture base materials provide a high shining finish to acrylic

areas that are hard to polish and reduce the adhesion of plaque or food residues [39,40]. Vallittu [41] tested the effect that surface treatments of auto-polymerising denture base material would have on the residual monomer content and release from the material. The test samples were either conventionally polished or treated with a light-cured resin. The results indicated that both surface treatment procedures were effective in reducing the content and release of residual monomer from the material, with the light-cured resin technique proving to be most effective. These results supported those of Szabó et al [42] who tested the effect of a light-cured resin on the properties of both auto- and heat-polymerised denture acrylic. The results indicated that the application of the light-cured resin may reduce the quantity of soluble components leaking from the material. The coated materials also recorded an increase in hardness, but this effect was thought to be countered by the increase in water sorption observed.

CONSTITUTION OF ENVIRONMENT

Denture base materials are exposed to a variety of substances in the oral cavity during function. For comfort, many patients prefer to sleep without their prosthesis, soaking it in water over night. Literature suggests that the molecular composition of the substance to which the material is exposed may affect the level of sorption and solubility experienced. As denture base materials are permanently exposed to fluids, Saini et al [12] investigated the sorption and solubility of heat-cured and self-cured acrylic resins immersed in different solutions. The solutions included distilled water, artificial saliva, denture cleansing solution, a mixture of distilled water and denture cleaning solution and a mixture of artificial saliva and denture cleaning solution. The study indicated that the sorption rate of heat-activated PMMA denture base material was lower when it was stored in an artificial saliva solution in comparison to the other substances. This observation was not limited to heat-activated acrylic, as the sorption rate of the self-activated PMMA denture base material was also lower when it was stored in an artificial saliva solution in comparison to the other substances. It was therefore concluded that the rate of sorption of PMMA denture base material may be affected by the molecular structure of the solution in which it is immersed. These findings may also be attributed to the difference in solubility parameters between the solution and the material [7], as well as the varying degrees of viscosity of the various solutions [19]. Similar results were recorded by Zidan et al [43], who investigated the long-term sorption and solubility of zirconia-impregnated PMMA

nanocomposite in water and artificial saliva. Although the study did not specifically aim to compare the sorption and solubility values recorded in distilled water against the values recorded in artificial saliva, it was noted that the conventional heat-cured samples soaked in artificial saliva recorded lower sorption and solubility values than those soaked in distilled water.

CONCLUSION

The sorption and solubility of heat-cured PMMA has a negative effect on the properties of the material, particularly its strength, dimensional stability, and biocompatibility. These factors all directly affect the longevity, performance, and comfort of the prosthesis. In order to ensure long term function, particularly for the elderly and those of lesser financial means, methods to reduce and control the amount of sorption and solubility experienced by heat-cured PMMA must feature strongly in the fabrication process. Mixing ratios and polymerisation cycles have been identified as external variables that can negatively influence the rate of sorption and solubility experienced by denture base materials while certain surface treatment procedures as well as storing the material in artificial saliva solution may optimise the lifespan of the denture. This review aims to assist future researchers and dental professionals

to better understand the importance of adhering to the manufacture's recommendations regarding the selection of materials, the clinical importance of a "polished" denture, and the effects that the molecular composition of a storage medium may have on the sorption and solubility properties of a prosthesis.

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DECLARATION OF INTEREST

The authors have no competing interests. Furthermore, the authors declare that they have no financial or personal relationships that may have inappropriately influenced them in writing this article.

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