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THE RESIDUAL MONOMER IN DENTAL ACRYLIC RESIN AND ITS ADVERSE EFFECTS

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Abstract: Acrylic based resins are frequently used in daily dental practice. The most common use of the materials includes denture bases and denture liners, temporary crowns and orthodontic appliances. In the mouth, properties and functional efficiency of applied acrylic resins depend on internal factors related to the methods and conditions of polymerization and on external factors that are related to the environment in which the material is placed. Residual monomer, which is released as a result of interaction of both sets of factors is often associated with irritation, inflammatory and allergic reactions of oral mucosa. The aim of this paper is to review literature dealing with the conditions of polymerization and biodegradation of acrylic resins under certain conditions in the oral cavity and their impact on oral health (reviewed literature available on Medline database during the past two decades.)

Conclusion: Methods and conditions of acrylate polymerization, on the one hand, and properties of saliva, chewing and the presence of microorganisms in the oral cavity, on the other hand, can be considered responsible for the release of residual monomers.

Clinically significant events followed by redness and erosion of the oral mucosa, burning sensation and burning mucosa and tongue, may be due to the effects of released, potentially cytotoxic, residual monomers.

Keywords: acrylic resin, dentures, residual monomer, cytotoxicity, biodegradation, oral manifestations.

INTRODUCTION

Acrylic-based resins are frequently used in daily dental practice, as they are able to provide the essential properties and have necessary characteristics for their use in diverse functions. Polymethyl methacrylate (PMMA)–based acrylic resins are used for fabrication of various dental prostheses and denture liners, temporary crowns and orthodontic appliances. Acrylic resin bases of removable partial or complete dentures and tooth-supported or implant-retained overdentures are used to replace the lost tissues and transfer masticatory forces from the denture to the residual ridges. Denture liners are used to improve the fit of denture bases, thus re-establishing the retention, support and stability of removable prostheses. Temporary crowns are used during the interval between tooth preparation and placement of the definitive crown, while orthodontic appliances are used for space maintenance, tipping teeth, overbite reduction, block movements and retention [1−5].

In the oral cavity, properties and functional values of acrylic resin based products depend on its endogenous factors caused by polymerization (degree of conversion of their constituent monomers, methods and the conditions of polymerization) [1-9] as well as exogenous factors caused by conditions present in oral cavity (saliva, bacteria, mastication) [5]. All these factors make a complex and intricate interplay of interactions, resulting in significant biological effect on oral cavity tissues. Biological, as the most common toxic effect on oral cells and tissues, achieves a residual monomer that occurs as a result of the polymerization process and/or biodegradation of dental materials in the oral cavity.

This article reviews the literature published during the past two decades, selected by use of a Medline search (US National Library of Medicine), which investigated residual monomer cytotoxic

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effects as a result of different polymerization methods and cycles and /or polymer biodegradation under certain conditions that are present in the oral cavity.

2. INFLUENCE OF THE METHODS AND CONDITIONS OF POLYMERIZATION ON THE RESIDUAL MONOMER CONTET IN DENTAL ACRYLIC RESINS AND ITS CYTOTOXICITY

Polymerization of a PMMA-based dental resin is an addition reaction that requires the activation of an initiator, such as benzoyl peroxide, which can then be decomposed by many different means, such as heat (heat polymerization) [1,3,4,6] or microwave polymerization [1,3,4,6,7] or by addition of a chemical activator, such as dimethyl-p-toluidine, at moderate temperatures (autopolymerization) [1−4,6,8] or light polymerization [1,3,4,6,9]. Polymerization is followed by conversion of methyl methacrylate (MMA) to PMMA (a curing process). During polymerization reaction of acrylic resins, not all the monomers are converted into polymers, and therefore some unreacted monomers called residual monomers are left. Its concentration varies depending on the methods and the conditions of polymerization [1−4,6−9]. Residual monomer is left in the polymer or might leach into water as well as human or artificial saliva [5,10−12]. Leached residual monomer is considered to be responsible for various degrees of in vitro cytotoxicity [10−15] and in vivo allergic responses [16−18].

The cytotoxic effect of denture base acrylic resins may be related to powder to liquid ratio, storage time, polymerization method, and cycle.

The polymer to monomer ratio is one of the variables that influence cytotoxicity of denture base acrylic resins. Jorge et al. [10] investigated the effect of polymer to monomer ratio on residual monomer levels and observed that resins prepared with a high proportion of polymer (5:3) resulted in significantly lower levels of residual monomer, as compared to those prepared with a lower ratio (4:3). Similarly, Kedjarune et al. [19] found that, the more monomer added to the mixture, the greater the amount of residual monomer and, therefore, the more potential for cytotoxicity.

Storage time is another feature that plays an important role in cytotoxicity of acrylic denture base materials. Sheridan et al. [20] reported that the cytotoxic effect of acrylic resins was greater in the first 24 hours after polymerization and that it decreased with time for all the resins evaluated in their study. The authors concluded that the longer the prosthesis is soaked, the less cytotoxic effect it is likely to have regardless of the denture base resin that it is made of. The cytotoxic effect may occur during several days after polymerization, but it can be minimized if the prostheses are stored in water for 24 hours [1,10,13,20]. It is hypothesized that the toxic substances released into the medium within the first 24 hours are either complexed with other chemicals in the medium or broken down over time that may alter their cytotoxic potential. Therefore, it is recommended that dentists soak the acrylic resin prostheses in water for at least 24 hours before placing them in the patient's mouth. It has been advocated that the prosthesis should be immersed in water at 50°C for 60 minutes, to reduce the amount of released monomer and hence the toxic potential of denture base resins, especially for autopolymerized resins. This is particularly important when hard autopolymerized reline resins are used [1,10,13,20]. The hypersensitivity reaction in the examined patients was decreased if prostheses were immersed in heated water. According to the authors, the decrease of the amount of residual monomer after this procedure may be due to further polymerization in the presence of free radicals. By immersing the prosthesis in heated water, monomer molecules diffuse more rapidly, reaching the remaining free radicals and leading to a complementary polymerization reaction. Similar results have been showed by Bural et al. [11] who have investigated the effect of post-polymerization heat-treatments on the degree of conversion, residual methyl methacrylate concentration and *in vitro* cytotoxicity of autopolymerizing acrylic repair resin. Authors concluded that post-polymerization heat-treatment of autopolymerizing acrylic repair resin by immersion in water at 60◦C for 30 min is clinically recommended to improve the degree of conversion while reducing the leaching residual MMA.

Depending on polymerization temperature and time, various quantities of residual monomer are left in the polymer resulting in different degrees of cytotoxicity. Kedjarune et al. [19] observed a reduced amount of residual monomer when polymerization time was extended, thus resulting in less cytotoxic effects. To define an ideal polymerization cycle for different acrylic resins, Harrison and Huggett [21] conducted a study where in 23 heat-polymerized denture base polymers were subjected to various polymerization cycles. The results of this investigation showed that a 7-hour incubation in water at 70°C followed by 1 hour at 100°C was ideal, because it provided maximum

conversion of residual monomer. In contrast, a 7-hour cycle at 60°C and the cycle of immersing the flask in boiling water, followed by a 5-minute immersion in water at 90°C, produced a high concentration of released residual monomers. Further, Bural et al. [12] conducted a study where 144 heat-polymerized denture base polymers were fabricated using 4 different polymerization cycles: (1) at 74°C for 9 h, (2) at 74°C for 9 h and terminal boiling (at 100°C) for 30 min, (3) at 74°C for 9 h and terminal boiling for 3 h, (4) at 74°C for 30 min and terminal boiling for 30 min. Specimens were eluted in a complete cell culture medium at 37°C for 1, 2, 5 and 7 days. Authors concluded that the use of terminal boiling in the polymerization process for at least 30 min and water storage of the heat-polymerized denture bases for at least 1 to 2 days before denture delivery is clinically recommended for minimizing the residual MMA and possible cytotoxic effects. These findings are in accordance with Bayraktar et al. [1] who concluded that the lowest overall residual MMA content was obtained from heat-polymerized specimens that were given a long-term terminal boil cure and then stored in distilled water at 37°C, for at least 1 day. For autopolymerized resins, the lowest residual MMA content was obtained when they were additionally polymerized in water at 60°C and then stored in distilled water at 37°C, for at least 1 day. Authors also concluded that at room temperature cured autopolymerized resins should be stored in water during at least 1 week. To assess the effect of polymerization time and temperature on the amount of residual methyl methacrylate monomer, Vallittu et al. [22] performed a study with 2 heat-polymerized resins activated by benzoyl peroxide and 2 autopolymerized resins in which the reaction was initiated by barbituric acid. The results showed that the heat-polymerized resins exhibited lower contents of residual methyl methacrylate compared to autopolymerized resins. This may be due to the rise of temperature in heat-polymerized resins, which resulted in mobility of the molecular chains, thereby facilitating the conversion of monomer into polymer. Thus, heating cycles with temperatures less than 100°C may result in polymers with higher methyl methacrylate contents than heating cycles with temperatures in excess of 100°C. It was also demonstrated that for autopolymerized resins, in which only the polymerization temperature was varied, the amount of residual monomer decreased as the temperature increased. Therefore, it seems reasonable to suggest that the autopolymerized acrylic resins should be heat-treated to decrease cytotoxic effects.

The method of polymerization is a decisive feature in the cytotoxicity of denture base acrylic resins. Ata and Yavuzyilmaz [23] have showed that cytotoxic effect is lower in heat-polymerized resins than in autopolymerized resins which is in accordance with the results of de Andrade Lima Chaves et al. [13] who have systematically reviewed the published literature on the cytotoxicity of denture base and hard reline materials. By reviewing literature, they have provided some evidence that heat-polymerized resins showed lower cytotoxic effects than autopolymerizing denture base acrylic resins and light-polymerized or dual-polymerized reline resins. The cytotoxic effects of chemically-activated, heat-activated, and microwave-activated acrylic resins on gingival fibroblasts were also reported by Sheridan et al. [20] who observed that, among the tested materials, the greatest cytotoxic effect was produced by chemically activated acrylic resins. Bayraktar et al. [1] revealed that autopolymerized resins eluted considerably more substances compared to the heat- and microwave-polymerized resins. These findings are in accordance with findings of Cimpam et al. [24] who also studied the effect of microwave heating on the residual monomer level of an autopolymerized resin used in the repair of prostheses. The results demonstrated that the specimens submitted to microwave irradiation after 20-minutes of autopolymerization showed a reduced amount of residual monomer when compared with resins undergoing other polymerization methods. A similar finding was observed by Blagojevic & Murphy [25] who showed that the residual monomer of an autopolymerizing resin decreased by approximately 4-fold when specimens were submitted to microwave irradiation. Therefore, it may be assumed that the reduction in residual monomer content by microwave irradiation could play an important role in decreasing the cytotoxic effects of autopolymerizing acrylic resins due to the heating that occurs. Azzari et al. [26] also reported a lower amount of residual monomer after microwave processing when this method was compared with the conventional heat-polymerizing technique. Thus, a shorter polymerization time and less residual monomer are considered as 2 of the advantages of microwave polymerization. Celebi et al. [4] compared the residual monomer release of acrylic denture base resins polymerized by hot water and microwave energy. Specimens polymerized by conventional methods exhibited slightly higher concentrations of residual monomer compared with specimens polymerized by microwave irradiation. The results from Bartoloni et al. [6], revealed that microwave irradiation yielded a substantial reduction of residual

monomer and a high degree of conversion of tested denture base acrylic resins, which is in accordance with findings of Bayraktar et al [1]. Visible light-polymerized denture base resins were introduced in the early 1980s. Although these resins have been reported to be nontoxic after polymerization [9], several studies have shown that these materials have varying levels of cytotoxicity [27,28]. The extent of their toxic effect appears to be related to the specific formulation of the material and polymerization time. Increasing the polymerization time may decrease resin toxicity [27]. Soaking prostheses fabricated with light-polymerized resins for 24 hours before insertion has been recommended to minimize exposure of oral tissue to cytotoxic substances such as methyl methacrylate and bis-GMA [29].

3. RESIDUAL MONOMER AS A CONSEQUENCE OF BIODEGRADATION

An important issue regarding the clinical application of acrylic based resins is their biodegradation. Changes of their chemical, physical and mechanical properties due to the oral environment conditions can be considered a definition of biodegradation. A major clinically significant consequence of acrylic based resins biodegradation is the producing of leachable, potentially toxic agents, most frequentlyresidual monomer, which in turn may induce a series of biological responses on cells and tissues. Polymer degradation does not occur as a result of isolated processes,as multiple factors as saliva, oral microbes and mastication, may be considered responsible for biodegradation processes [5,30].

Saliva, the product of small and large salivary glands, is composed of several components that may significantly contribute to biodegradation of the acrylic based resins [5]. Water is the most abundant component of saliva as such is one of the main factors to cause biodegradation. Water molecules can easily penetrate the polymer network allowing the diffusion of unbound/uncured monomers and/or additives from the material network [31,32]. There are two conditions that influence the amount of water diffusion to denture resins. One is the water diffusion coefficient of the material that affects the time needed for its saturation with water. The other is the amount of residual components that are released in the medium and replaced by water molecules [32,33].

Polymeric structures and dental materials in particular may also be chemically degraded in aqueous solutions essentially through two mechanisms: hydrolysis and enzymatic reaction [30-33]. Salivary enzymes can degrade polymers through attacks on the side chains, producing both potentially harmful by-products as well as a deterioration of the network properties. The composition of the monomers producing the network is a major factor in determining the extent of degradation, especially when enzymes are responsible. Various esterases that have been shown to be present in saliva can promote esterification of methacrylates [34,35]. The effect of enzyme degradation on mechanical properties has been manifested as a reduction in surface hardness and wear resistance [33−35].

Interactions between oral microbes and the polymer dental materials may also occur, suggesting some surface degradation effect caused by bacteria colonization (increasing in the roughness) [36]. Most microorganisms that are present intraorally, especially those responsible for caries, periodontal disease, and denture-related stomatitis, can only survive in the mouth if they adhere to non-shedding oral surfaces and start forming colonies [37]. Bacterial adhesion on hard dental surfaces is followed by the accumulation of dental plaque [36]. Surface roughness and the surface free energy play a key role during this process [37,38]. Changes in these clinically important variables might have a significant influence on bacterial adhesion and retention [38]. Surface free energy varies for different dental materials. A thin biofilm of the acquired salivary pellicle can significantly reduce free energy on hard intraoral surfaces. Several studies have demonstrated that rough acrylic resin surfaces are significantly more prone to bacterial accumulation and plaque formation than smooth surfaces [37−39]. The findings of Quirynen et al. [38] indicated that supragingivally the impact of surface roughness on microbial adhesion is much more important than the influence of surface free energy.

Biodegradation of the materials in the oral cavity can also be induced by fatigue, which is caused by relatively weak repetitive loads such as ordinary, masticatory force. A continuous application of mechanical and environmental loads leads to progressive degradation and crack initiation and growth, resulting in catastrophic failure of the resins. This process is further assisted by pre-existing voids introduced during the material processing and residual stresses [40,41]. Mastication can also apply shear and compression forces on denture teeth causing wear. [41].

The release of compounds from different types of acrylic based resins, as a consequence of

biodegradation, has been widely investigated. Mostly, investigations have been conducted in the experimental conditions (incubating polymer specimens of different shapes and sizes prepared according to manufacturer's instructions in a liquid, at room temperature or 37 °C, for periods of time ranging from hours to 1 or 2 months). Water was used as the leaching media [1−3,10−13,15,33,42]. In some studies [2,16,20,42], the diffusion of residual monomers and other leachable components from acrylic based materials in human and artificial saliva has been investigated. Kedjarune et al. [19] used unstimulated whole human saliva to evaluate the release of MMA from heat-cured and autopolymerized resins. Very few investigators have concentrated on evaluating the release of compounds from acrylic based materials in clinical studies [16,43]. Tsuchiya et al. [43] found significant amounts of formaldehyde and MMA in human saliva under *in vivo* conditions leaching from acrylic autopolymerized resins. Further, Goncalves et al. [16] evaluated the *in situ* levels of residual MMA monomer of an autopolymerized acrylic resin in forty volunteers. High concentrations of residual monomer during the first 24h of use were observed. In spite of different methodologies the majority of published studies refers to elution of unbound components, mainly MMA monomer [2,3,10,12,15,20,33,42], as one of the main consequences of material biodegradation.

Leaching of the residual monomer may influence biocompatibility of denture material. Products of acrylic based resins biodegradation have been suspected of being a contributing factor for local chemical irritation, sensibilization and pain, labial edema, mucosal inflammation or ulceration, oral diseases such as a burning mouth syndrome and denture stomatitis, systemic allergic reactions due to acrylic resin. There is an assumption that residual monomers in the denture base which is in continuous contact with great part of oral mucosa, might have clinically affected the surrounding tissues [16−18,44,45). In addition to these symptoms, many studies have focused on the cytotoxicity of leached MMA monomer [11−15,19,20,23,24,27−30,32,43). Test systems vary considerably in the way cytotoxicity is measured but all indicate changes in basic cell structures, such as cell membrane integrity and cell functions like enzyme activities or the synthesis of macromolecules [46]. The mechanism of adverse effect caused by MMA monomer is thought to involve direct toxicity from released or residual MMA and oxidative stress created by free radicals that are released during the resin polymerization [46,47].

The results of cytotoxicity tests are limited in the sense of their applicability to their clinical use. The gap that exists between the results published by research laboratories and clinical reports should be shortened. Further well-controlled clinical studies are necessary to improve the knowledge of materials biocompatibility in intraoral conditions.

4. CONCLUSION

On the basis of the review of literature, it may be concluded that:

1. Acrylic-based resins are intensively used in dentistry practice as denture base materials, liners, restorative or orthodontic appliances materials. These substances are made by polymerization of methacrylate related monomers.
2. Increasing concern arises regarding safe clinical application of these materials due to methods and conditions of polymerization and their biodegradation under the oral environment.
3. Concerning the methods and the conditions of polymerization, cytotoxic effect of denture base acrylic resins may be related to powder to liquid ratio, storage time, polymerization method, and cycle.
4. Causes for biodegradation comprise several factors such as saliva characteristics, mastication and oral microbes.
5. Consequences of polymerization process and materials biodegradation refer mainly to the release of potential cytotoxic compounds from the polymer network with different adverse effects on oral health (irritation, inflammation, and an allergic response of the oral cavity).
6. There is an opportunity for future research in different areas related to the evaluation of acrylic based resins polymerization and biodegradation. This will lead to a more concise definition of biocompatibility issues related to these dental materials.

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РЕЗИДУАЛНИ МОНОМЕР АКРИЛАТНИМ СМОЛАМА У СТОМАТОЛОГИЈИ
И ЊЕГОВИ НЕГАТИВНИ ЕФЕКТИ

Сажетак: Акрилатне смоле као градивни материјали често се употребљавају у свакодневној стоматолошкој пракси. Најчешће се користе за израду базе зубних протеза те као лајнери за подлагање протезне базе, за израду привремених круница и ортодонтских апарата. У усној дупљи, особине и функционална ефикасност примијењених акрилатних смола зависе како од унутрашњих фактора који се односе на методе и услове полимеризације тако и од спољашњих који су односе на услове средине у коју се материјал пласира. Резидуални мономер, који се ослобађа као посљедица интеракције обје скупине фактора често се доводи у везу са појавом иритација, упалних и алергијских реакција слузнице усне шупљине.

Циљ рада је преглед литературе која се бавила условима полимеризације те биоразградњом акрилатних смола под одређеним условима те њиховим утицајем на орално здравље (прегледана литература доступна на Medline бази података у посљедње двије деценије).

Закључак: Методе и услови полимеризације акрилата, са једне стране, те особине пљувачке, мастикација и присуство микроогранизма у усној дупљи, са друге стране, могу се сматрати одговорним за ослобађање резидуалног мономера.

Клинички значајне манифестације праћене црвенилом и ерозијом оралне слузнице, осјећајем печења и жарења слузнице и језика, могу се јавити као посљедица дејства ослобођених, потенцијално цитотоксичних, резидуалних мономера.

Кључне ријечи: акрилатне смоле, зубна протеза, резидуални мономер, цитотоксичност, биоразградња, оралне манифестације.

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