

Loss of residual monomer from resilient lining materials processed by different methods

Perda de monômero residual de reembasadores resilientes processados por diferentes métodos

Abstract

Purpose: To evaluate residual monomer release from resilient denture lining materials polymerized by different methods.

Methods: Two materials were assessed: Ever-Soft polymerized by hot water bath or microwave energy, and Light Liner polymerized by chemical reaction and visible light (dual polymerization). Residual monomer release was measured in 12 specimens (40x10x0.3mm) made of each material and polymerization method. The specimens were stored in distilled water for 168 hours at 37°C, and analyzed daily by ultraviolet spectrometry (Light Liner: 204nm, Ever-Soft: 206nm). Data were analyzed by one-way ANOVA and Bonferroni test ($\alpha=0.05$). The residual monomer released as a function of time was determined by polynomial regression analysis.

Results: Residual monomer amount released at 168 hours from specimens polymerized by hot water bath ($0.27 \pm 0.01 \mu\text{g}/\text{cm}^2$) was significantly greater ($P < 0.05$) than from those processed by microwave energy ($0.25 \pm 0.02 \mu\text{g}/\text{cm}^2$). Ever-Soft showed decrease in monomer release over time, tending to stabilize at 96 hours. Light Liner continued to release monomer over time.

Conclusion: Ever-Soft may be polymerized by microwave energy. Residual monomer release values were low, and the monomer levels decreased over time.

Key words: Laboratory research; resilient lining material; residual monomer

Resumo

Objetivo: Avaliar o monômero residual liberado de materiais resilientes para reembasamento polimerizados por diferentes métodos.

Metodologia: Dois materiais foram testados: Ever-Soft polimerizado por banho quente de água ou por energia de microondas, e Light Liner polimerizado quimicamente e por luz visível (polimerização dual). O monômero residual liberado foi mensurado em 12 espécimes (40x10x0,3mm) fabricados com cada material e método de polimerização. Os espécimes foram armazenados em água destilada por 168 horas a 37°C, e analisados diariamente por espectrometria ultravioleta (Light Liner: 204nm, Ever-Soft: 206nm). Os dados foram analisados por ANOVA de fator único e teste de Bonferroni ($\alpha=0,05$). O monômero residual liberado em relação ao tempo foi determinado por análise de regressão polinomial.

Resultados: O monômero residual liberado em 168 horas do teste ($\mu\text{g}/\text{cm}^2$) em espécimes polimerizados por banho de água quente ($0,27 \pm 0,01 \mu\text{g}/\text{cm}^2$) foi significativamente maior ($P < 0,05$) que em espécimes processados por energia de microondas ($0,25 \pm 0,02 \mu\text{g}/\text{cm}^2$). Ever-Soft mostrou uma redução na liberação de monomer residual com o tempo, tendendo a se estabilizar em 96 horas. Light Liner continuou a liberar monômero com o tempo.

Conclusão: Ever-Soft pode ser polimerizado por energia de microondas. Os valores de monômero residual liberado foram baixos, e os níveis de monômero diminuíram com o tempo.

Palavras-chave: Estudo laboratorial; reembasador resiliente; monômero residual

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Introduction

The success of removable partial prostheses depends on several factors, such as comfort, function, and esthetics. However, in view of continual bone resorption, these prostheses may become maladapted (1), leading to alterations to bearing tissues and consequently affecting masticatory function. Resilient denture lining materials can be used to allow patient comfort and distribute masticatory forces more homogeneously (2,3). These materials can be classified according to their chemical composition into acrylic resin-based and silicone-based materials (4). The acrylic resin-based denture liners can be divided into two subgroups. The first comprises materials in which the liquid is made of monomer components, such as methyl, ethyl or butyl methacrylate and plasticizing agents. The second group is similar to tissue conditioners, in which the liquid contains a mixture of plasticizer esters and ethyl alcohol (4,5).

Resilient denture lining materials can be processed in different ways and are classified as chemical, heat- or light-activated. For heat-activated materials, heat can be generated by hot water bath (6,7) or microwave energy (8,9), while the light-activated liners use visible light as energy source (7,10,11). The visible light polymerization method presents low dimensional alteration (7), adequate bond strength to denture bases (12,13), complete polymerization without residual components, and absence of methylmethacrylate in their chemical composition (14). On the other hand, microwave energy irradiation is considered to be faster and cleaner compared with the hot water bath method (8). Another advantage of this method is the control of polymerization time (9) and low release of residual monomer (15,16).

Several researchers have analyzed residual monomer release from acrylic resins, as this property relates to the degree of material polymerization (15,17-20) and influences several physical and mechanical polymer properties (21). However, data on resilient denture liners are scarce (14,22). Thus, the aim of this study was to compare the residual monomer (methylmethacrylate and butylmethacrylate) release from two resilient denture lining materials, polymerized by hot

water bath, microwave energy or dual polymerization (chemical and visible light) after seven days of storage in distilled water.

Methodology

The resilient lining materials used in this study and their chemical composition are listed in Table 1. Twelve rectangular specimens, 40x10x0.3mm, were fabricated with Ever-Soft and Light Liner resilient material, in accordance with the manufacturers' instructions. Twelve additional specimens were prepared with Ever-Soft using microwave energy irradiation. All Ever-Soft specimens were obtained by investing rectangular silicone matrices (Optosil P Plus, Heraeus Kulzer GmbH, Hanau, Germany) in type III dental stone (Herodent Soli-Rock, Vigodent, Rio de Janeiro, RJ, Brazil) using a standard metal dental flask (Uraby, DLC, São Paulo, SP, Brazil) or plastic flask (Onda-Cryl, Artigos Odontológicos Clássico Ltd., São Paulo, SP, Brazil) for hot water bath and microwave polymerization, respectively. The mold cavities were filled with Ever-Soft and polymerized by hot water bath, in accordance with the manufacturers' instructions (40°C for 15min, followed by 60°C for 45min), and by microwave energy (600W for 4min), as previously described (23). All flasks were bench cooled for 2 hours before opening. The specimens were removed and finished using progressively smoother aluminum oxide papers (320-, 400-, and 600-grit) in a horizontal polisher (Model APL-4, Arotec, São Paulo, SP, Brazil). After finishing, two layers of sealer (Myerson, Austenal Inc, Chicago, IL, USA) were applied over the entire surface of the Ever-Soft specimens, with a 15- min interval between applications.

Light Liner specimens were prepared using a transparent matrix (23) and polymerized in a light box (EDG Lux, EDG Equipamentos e Controles, São Paulo, SP, Brazil) for 5min. After polymerization, an air barrier coating (Harry J. Bosworth, Skokie, IL, USA) was applied on the specimen surface to avoid formation of an air-inhibited layer. The specimens were again polymerized for 5min as recommended by the manufacturer. Polishing procedures were carried out as previously described.

Table 1. Resilient denture relining materials used in this study

Resilient Liners	Composition	Processing Method	Manufacturer
Ever-Soft	Powder: Polyethyl methacrylate Liquid: Dibutyl phthalate, ethyl alcohol, ethyl acetate Sealer: Methyl ethyl ketone	Chemical or hot water bath	Myerson, Austenal, Inc, Chicago Ill, USA
Light Liner	Powder: polyethyl methacrylate, benzoyl peroxide Liquid: n-butyl methacrylate, butyl plasticizer, ethoxylated, bisphenol dymethacrylate, dymethacryl-p-tolouidine, camphorquinone. Air barrier: polyester polyol	Dual polymerization (chemical and visible light)	Harry J. Bosworth, Skokie, Ill, USA

Residual monomer was assayed using a modified technique from Lamb et al. (17). Each specimen was placed in a small assay glass tube (Pyrex, Corning Life Sciences, Acton, MA, USA) containing 6.6mL of deionized water. The test tubes were sealed with parafilm (Parafilm M, Pechiney Plastic Packaging, Menasha, WI, USA) and stored at $37^{\circ}\text{C}\pm 2$ for 24 hours. After incubation, the aqueous solution was removed and analyzed. The release of methylmethacrylate from Ever-Soft material was analyzed at 206nm in a spectrophotometer (DU-70, Beckman, Fullerton, CA, USA) to determine residual monomer levels. The butyl methacrylate released from Light Liner was analyzed at 204nm using the same equipment. The aqueous solution was discharged after each reading. Each specimen was washed in distilled water, dried with absorbent paper, and replaced in the test tube previously filled with deionized water. It was important that the water filled the assay tube completely because monomer is sensitive to oxygen, and levels of monomer release decrease in water storage. Spectrophotometric analyses were carried out daily for 7 days, until stabilization of residual monomer release.

The accuracy of the calibration curve for Ever-Soft was determined by preparing 1% v/v solutions of methylmethacrylate (SIGMA, lot: 56H3407) in deionized water ($2.36\text{-}18.86\text{mg/mL}^{-1}$). The calibration curve for Light Liner was determined by means of 1% v/v solutions of butyl methacrylate ($2.23\text{-}17.88\text{mg/mL}^{-1}$, Aldrich, lot: 07426AO) in 50% ethyl alcohol. The calibration curves were prepared 24 hours before readings were taken, so that they would have the same time period and storage conditions as the test specimens at 37°C . The specimen size was calculated, and the results were expressed in mg of monomer released per cm^2 of specimen area.

Methyl methacrylate release data were analyzed by one-way analysis of variance (ANOVA), and the means were compared using the Bonferroni test ($\alpha=0.05$). The release of methyl methacrylate and butyl methacrylate over time was analyzed by polynomial regression analysis.

Results

The means of methylmethacrylate residual monomer released by Ever-Soft material polymerized by microwave energy and hot water bath methods are displayed in Table 2. For Ever-Soft, methylmethacrylate levels were significantly different between polymerization methods only after 168 hours ($P<0.05$). Specimens polymerized by microwave energy had lower monomer release ($0.25\pm 0.02\mu\text{g/cm}^2$) than those polymerized by hot water bath ($0.27\pm 0.01\mu\text{g/cm}^2$). The amount of methylmethacrylate released in water over time from Ever-Soft is shown in Figure 1, and the data of butyl methacrylate released from Light Liner are presented in Figure 2. Both materials showed that residual monomer release was high during the first days (hot water bath: $0.62\pm 0.02\mu\text{g/cm}^2$, microwave energy: $0.66\pm 0.05\mu\text{g/cm}^2$, visible light: $1.04\pm 0.02\mu\text{g/cm}^2$) but decreased over time. Residual monomer release from Ever-Soft tended to stabilize after 96 hours (hot water bath: $0.30\pm 0.02\mu\text{g/cm}^2$

and microwave energy: $0.29\pm 0.03\mu\text{g/cm}^2$). In contrast, Light Liner continued to release monomer over time.

Table 2. Mean* residual monomer content ($\mu\text{g/cm}^2$) of Ever-Soft polymerized by hot water bath and microwave energy methods

Time (h)	Processing Method	
	Hot water bath ($\mu\text{g/cm}^2$)	Microwave energy ($\mu\text{g/cm}^2$)
24	0.62 ± 0.02 A	0.66 ± 0.05 A
48	0.41 ± 0.02 A	0.41 ± 0.04 A
72	0.39 ± 0.01 A	0.40 ± 0.04 A
96	0.30 ± 0.02 A	0.29 ± 0.03 A
120	0.30 ± 0.01 A	0.30 ± 0.04 A
144	0.22 ± 0.01 A	0.22 ± 0.03 A
168	0.27 ± 0.01 B	0.25 ± 0.02 A

* Mean values followed by different letters are statistically different ($P<0.05$)

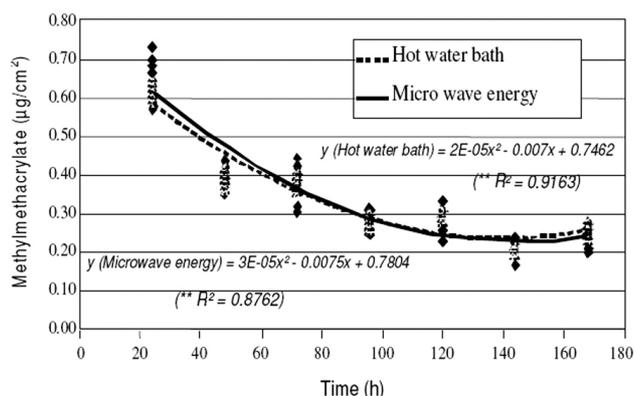


Fig. 1. Polynomial regression to evaluate the effect of time on release of methyl methacrylate from Ever-Soft resilient denture lining material.

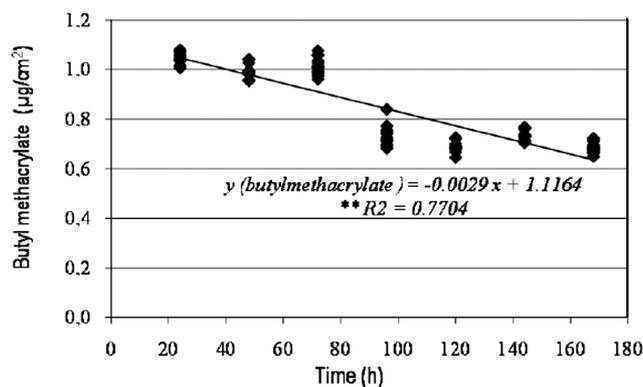


Fig. 2. Polynomial regression to evaluate the effect of time on release of butyl methacrylate from the Light Liner resilient denture lining material.

Discussion

Resilient lining materials are widely used for lining removable partial prosthesis, providing adaptation and comfort to patients with alterations to support tissues (1). When immersed in aqueous medium, resilient liners undergo water sorption and/or simultaneous leach of plasticizers and other soluble components, such as ethyl alcohol (4). This study assessed the release of two types of residual monomer from resilient denture lining materials processed by different methods, after seven days of storage in distilled water.

There was no difference in the values of methylmethacrylate released from Ever-Soft polymerized by microwave energy and hot water bath from 24 to 144 hours (Fig. 1). However, a significant decrease in residual monomer release was observed in Ever-Soft specimens polymerized by microwave energy after 168 hours. It is possible that the material polymerized by hot water bath and microwave energy reached the same monomer conversion rate in the time interval from 24 to 144 hours of assessment (20). At 168 hours, greater diffusion of the non-reacted monomer at the surface of the denture liner polymerized by microwave energy may have occurred. Consequently, it was volatilized, and there was a reduction of monomer release in water (16). Residual monomer release values were higher in the first days and decreased over time for all polymerization methods as previously shown (15,17). After polymerization, monomer levels may decrease by two mechanisms: i) monomer diffusion from the resin bulk into the water medium; or ii) additional polymerization of the same material, as active radicals were detected among the chains that promote monomer polymerization (17).

The type of residual monomer seemed to affect release rates in water. Methylmethacrylate release from Ever-Soft polymerized by microwave energy and hot water bath was stabilized after 96 hours, but butyl methacrylate released from Light Liner continued to decrease over time. Probably, butyl methacrylate has less solubility in water, which may slow down the cross linking process with the polymer (14). Clinically acceptable values for residual monomer release range from 1 to 3% (1). In the present study, methylmethacrylate release ranged from 0.22 $\mu\text{g}/\text{cm}^2$ to 0.66 $\mu\text{g}/\text{cm}^2$ and butyl methacrylate from 0.68 $\mu\text{g}/\text{cm}^2$ to 1.04 $\mu\text{g}/\text{cm}^2$. These results are different from those found by Kawaguchi et al. (14), who reported a figure of 2.32%, and may be due to differences in baseline monomer concentrations in denture lining materials (22), polymerization methods, storage time, and assessment method.

The findings of this study helped to determine that Ever-Soft resilient lining material can be polymerized by microwave energy with satisfactory results. Light Liner material presented low residual monomer release values. Thus, further studies should assess monomer release from other resilient materials, as this property may alter several physical properties of resilient denture lining materials, such as bond strength to denture bases, hardness, and permanent deformation.

Conclusions

Within the limitations of this *in vitro* study, it was concluded that Ever-Soft resilient lining material can be polymerized by microwave energy. Residual monomer levels released from Ever-Soft and Light Liner resilient lining materials were low and decreased over time.

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