




COLOR VARIATIONS IN PMMA ACRYLIC RESINS AFTER THERMOCYCLING AND MOUTHWASH SIMULATION

Variações de cor em resinas acrílicas de PMMA após termociclagem e simulação de enxaguatório bucal

Access this article online	
Quick Response Code:	Website: https://periodicos.uff.br/ijosd/article/view/61091
	DOI: 10.22409/ijosd.v1i66.61091

Autores:**Fabiano Martins Malafaia**

Professor Adjunto das Disciplinas de Prótese Total Removível e Estágio Supervisionado Clínica do Adulto II da Faculdade de Odontologia da Universidade Federal Fluminense. Departamento de Odontotécnica (MOT), Niterói-RJ, Brasil.

Carlos Alexandre Lopes Marques

Professor Adjunto das Disciplinas de Prótese Total Removível, Estágio Supervisionado Clínica do Adulto I e II e Odontologia Digital da Faculdade de Odontologia da Universidade Federal Fluminense. Departamento de Odontotécnica (MOT), Niterói-RJ, Brasil.

José Carlos Magalhães

Professor Adjunto das Disciplinas de Prótese Fixa e Estágio Supervisionado Clínica do Adulto I da Faculdade de Odontologia da Universidade Federal Fluminense. Departamento de Odontotécnica (MOT), Niterói-RJ, Brasil.

Flávio Henriques Queiroz

Professor Adjunto das Disciplinas de Prótese Total e Odontologia Digital da Faculdade de Odontologia da Universidade Federal Fluminense. Departamento de Odontotécnica (MOT), Niterói-RJ, Brasil.

Amanda de Oliveira Pinto Ribeiro

Mestre em Odontologia Restauradora – Universidade Estadual Paulista- Instituto de Ciência e Tecnologia de São José dos Campos (ICT-SJC).

Endereço para correspondência: Fabiano Martins Malafaia Rua : Estrada dos Três Rios, 1619 apt 401 Freguesia.

E-mail para correspondência: fmalafaia@gmail.com

RESUMO

Estabilidade de cor é um requisito estético fundamental das resinas acrílicas provisórias, em especial quando esses materiais são submetidos a longos períodos na cavidade oral. Embora resinas acrílicas temporárias novas e aprimoradas estejam disponíveis, os efeitos da variação térmica e de soluções antissépticas na estabilidade de cor de resinas acrílicas provisórias ainda não foram completamente elucidados. Portanto, este estudo avaliou a coloração de resinas autopolimerizáveis polimetilmetacrilato (PMMA) e bis-acrílico submetidas à termociclagem e imersão em clorexidina. Amostras padronizadas (n=10) foram preparadas de duas resinas acrílicas temporárias a base de PMMA (Alike® e Duralay®). Foram realizadas três avaliações de cor (T1 – 24h após o preparo da amostra, T2 – após termociclagem e T3 – após termociclagem e imersão em clorexidina) por meio de espectrofotômetro eletrônico de seleção de sombra (Vita Easy Shade). Os dados obtidos foram analisados por ANOVA e teste t ($\alpha=0,05$). As resinas de PMMA Alike e Duralay apresentaram alteração de cor após a termociclagem e também após a imersão em clorexidina.

Palavras-chave: Estabilidade de cor; resinas acrílicas; termociclagem; clorexidina.

ABSTRACT

Color stability is a fundamental aesthetic requirement for temporary acrylic resins, especially when these materials are subjected to long periods in the oral cavity. Although new and improved temporary acrylic resins are available, the effects of thermal variation and antiseptic solutions on the color stability of temporary acrylic resins have not yet been fully elucidated. Therefore, this study evaluated the color of self-polymerizing resins polymethyl methacrylate (PMMA) and bis-acryl subjected to thermocycling and immersion in chlorhexidine. Standardized samples (n=10) were prepared from two PMMA-based temporary acrylic resins (Alike® and Duralay®). Three color evaluations were carried out (T1 – 24h after sample preparation, T2 – after thermocycling and T3 – after thermocycling and immersion in chlorhexidine) using an electronic shade selection spectrophotometer (Vita Easy Shade). The data obtained were analyzed by ANOVA and t test ($\alpha=0.05$). PMMA Alike and Duralay resins showed color changes after thermocycling and also after immersion in chlorhexidine.

Key-words: Color stability; Acrylic Resins; Thermocycling; Chlorhexidine.



INTRODUCTION

Temporary resin restorations must provide function, comfort and aesthetic appeal to the prepared tooth structure. Still, provisional restorations are used for diagnostic purposes and treatment planning, mainly to correct irregular occlusion, vertical dimension, changes in the location and contour of the gingival margin, size, shape and color of the final restoration (CHRISTENSEN, G 2003 e HASLTON et al., 2005). Therefore, in complex and prolonged treatments, provisional restorations are an essential part of the treatment process and the integrity of these restorations must be preserved throughout the diagnostic and restorative phases. Consequently, discoloration of these resins is critical and concerning as it can lead to patient discontent, additional chair time, and expense for replacement (GULER et al., 2005).

Three types of chemically polymerized materials are commercially available for provisional restorations of single and multiple dental elements: polyethylmethacrylates, polymethylmethacrylates, and bisacrylic resin composites (CHRISTENSEN, G 2003). Due to their improved clinical characteristics, methylmethacrylates and bisacrylic resin are generally the most common temporary materials used in practice dental (CHRISTENSEN, G 2003). Although these acrylic resins have clinical advantages over ethyl methacrylate resins, provisional acrylic resins have low resistance to wear, abrasion, impact and color stability (CHRISTENSEN, G 2003; RUTKÚNAS et al., 2010; ALVES et al., 2007; SILVA et al., 2006).

The discoloration of self-polymerizing acrylic resins is mainly promoted by extrinsic factors, such as staining food and drinks (ZIMMERLI et al., 2011). In clinical oral rehabilitation, chlorhexidine is routinely used and is an effective antimicrobial for mouthwash, (SILVA WC et al., 2007) but this antiseptic agent is capable of promoting undesirable effects, such as, tongue sensitivity and acquired pellicle coloration leading to darkening of the enamel. Additionally, prolonged use of chlorhexidine can lead to discoloration of temporary crowns and bridges (SILVA et al., 2007; CAL E et al., 2007).

In extensive prosthetic rehabilitations, temporary crowns and bridges manufactured with self-polymerizing temporary resins must be resistant and minimum aesthetic requirements to become clinically effective. However, little is known about the behavior of these materials when aged in the oral cavity and subjected to chlorhexidine staining. Therefore, the objective of this study was to evaluate in vitro the effects of thermocycling and 0.12% chlorhexidine solution on the color stability of two conventional methyl methacrylate (PMMA) temporary acrylic resins. The alternative hypothesis tested (H_a) is that thermal variation and



immersion in chlorhexidine will promote color changes of the supply resin-based materials.

MATERIAL AND METHODS

Sample preparation and 1st color assessment (T1)

The samples were prepared in a stainless steel mold (25 mm x 10 mm and 2.5 mm wide) filled in a single increment with temporary acrylic resins (n=10) of each methyl methacrylate (PMMA) resin (Alike® – GC American, Alsip, IL, USA and Duralay® – Reliance Dental Mfg. Co, Worth, IL, USA) (Table 1). The PMMA resins (Alike® and Duralay®) were handled according to the manufacturer's instructions and inserted into the mold using disposable syringes. The material was adapted by compressing a polyester strip under a glass plate, with a static load of 1.5 kg, to remove excess material. The samples were polymerized in a short-cycle water bath, following the manufacturer's instructions (VH Equipamentos, São Paulo, Brazil), 74°C for 1 hour and 100°C for 30 min. The samples were also finished and polished with HF079FE-060 tungsten burs (Brightdent NTI, Kahla, Germany). Then, were immersed in a remineralizing solution according to the solution proposed by (SHINKAI RS et al., 2001) at 37°C, composed by 1.5 mM CaCl₂, 0.9 mM Na₃PO₄, 0.15 M KCL, pH 7.0, 3.125 mL/mm², to simulate the clinical picture. The manufactured samples were kept in artificial saliva for 24 hours, removed, carefully air-dried, stored in a Pasteur oven at 37°C for 2 hours and subjected to the first color assessment (T1). The initial color assessment was determined after immersion in a remineralizing solution to simulate the clinical picture and standardize the humidity of all samples.

Thermocycling and 2nd color assessment (T2)

Samples from all groups were subjected to thermocycling (MSCT-3 PLUS - Marcelo Nucci-ME, São Carlos, Brazil) in a deionized water bath for 15 s, at 50 - 55o ± 1oC, to age the samples, simulating temporary crowns. Acrylic and bridges subjected to prolonged oral rehabilitation. The groups performed 2,000 cycles and were stored in a remineralizing solution for 24 hours after the thermocycling procedure. The samples were then completely air-dried, stored in a Pasteur oven at 37oC for 2 hours and subjected to a second color assessment (T2 – color assessment after thermocycling). Subsequently, the samples were kept in a remineralizing solution for 24 hours and immersed in a 0.12% chlorhexidine solution.



Immersion in 0.12% corhexidine solution and 3rd color assessment (T3)

The provisional acrylic resin samples were individually immersed in 5 ml of 0.12% chlorhexidine (Noplak, Daudt, São Paulo, Brazil) at 37°C, for 1 min under agitation. Subsequently, the samples were rinsed and returned to the remineralizing solution. This procedure was performed twice a day (8 am and 6 pm), for 14 days, with the aim of simulating the clinical protocol (SILVA WC et al., 2007). Afterwards, the samples were completely air-dried, stored in a Pasteur oven at 37°C for 2 h, and subjected to the third color measurement (T3).

Color measurement

Color measurement was carried out using an electronic shade selection spectrophotometer (Easy Shade, Vita – Germany) at each experimental time (T1, T2, T3) which allowed color determination using the CIE color evaluation system (Standard Commission Internationale de L'Éclairage Color System). According to this system, color can be determined based on three coordinates (L^* , a^* and b^*) that quantify the three dimensions of the color of a specific object. The " L^* " value corresponds to brightness (the value 100 refers to perfect white color and 0 refers to black); " a^* " determines the amount of red (positive values) and green (negative values); " b^* " is related to the amount of yellow (positive values) and blue (negative values) (ASSUNÇÃO e SOUZA et al., 2006).

The device was calibrated before each measurement, as recommended by the manufacturer. Measurements were carried out perpendicular to the surface of each sample and two readings were taken on each sample at each experimental time and the average value was obtained and considered for statistical analysis.

The values of the coordinates L^* , a^* , b^* were obtained and the discoloration of the provisional acrylic resins in the three evaluation periods (T1, T2 and T3) was expressed according to equation (Friele-Mac Adam-Chickering): $\Delta E = (\Delta L^2 + \Delta a^2 + \Delta b^2)^{1/2}$, where L^* , a^* and b^* represent the difference of the three readings performed on each sample. The resulting value obtained (ΔE) is the color difference between the experimental evaluation periods. According to (VICHI A et al., 2004), $\Delta E = 3.3$ is the limit for clinical acceptance of the color of a composite material.

Statistical analysis

The color measurement data in the three experimental periods were statistically analyzed. Normal distribution was verified by the Kolmogorov-Smirnov and Lilliefors tests ($p > 0.05$) and ANOVA and t-test ($p < 0.05$) were performed with a significance limit set at 5% using the BioEstat 2.0 software (Instituto Mamirauá ,

CNPq, Belém do Pará, Brazil). The color variation of the acrylic resin was evaluated over time, but no comparisons were made between the acrylic resins, as even though they have the same shade (B3), each acrylic resin has different color values.

RESULTS

Average values and standard deviations are presented in Table 1. Higher numerical values represent greater discoloration of acrylic resins.

Table 1. Means and standard deviations of discoloration of acrylic resins at each experimental time (T1 – 24h after sample preparation, T2 – after thermocycling and T3 – after thermocycling and immersion in chlorhexidine).

Resins – Basic composition	Color evaluation		
	T ₁	T ₂	T ₃
Alike ¹ (Methyl-methacrylate, ethyl -2-cyano 3-3 diphenylacrylate, methanol, barium silicate glass, benzoyl peroxide, dibutylphthalate).	0.37(0.16) a	1.47(0.96) b	2.97(0.67) c
Duralay ² (Methyl-methacrylate, polymer, dialkylphthalate, benzoyl peroxide)	1.30(0.53) a	3.05(0.89) b	4.22(0.84) c

¹GC American, Alsip, IL, USA; ² Reliance Dental, Mfg. Co, Worth, IL, EUA.

The results showed a change in the color of PMMA acrylic resins (Alike® and Duralay®) between T1 and T2 ($p=0.0084$). Resin materials showed color change after thermocycling (T2, $p<0.05$), Chlorhexidine was able to intensify the color change of PMMA resins (T3, $p=0.0009$) (Alike® and Duralay®) in compared to T2.

DISCUSSION

Autopolymerized resins are more prone to discoloration than thermopolymerized temporary acrylic resins. This is due to the delay in the oxidation process of tertiary amines, which are the molecules that initiate the polymerization reaction (PHOENIX, RD 1996). Furthermore, the use of an oral antiseptic can increase the coloring capacity of temporary acrylic resins (CAL E et al., 2007; SEDANUR et al., 2013).



It has been previously reported that the artificial aging process promoted by thermocycling methods interferes with the durability of composite resins, as it can dislodge inorganic particles from the organic matrix, (SEDANUR et al., 2013) promote microcracks at the organic/inorganic interface and surface tensions due to the high degree of temperature variation. close to the surface (KAWANO et al., 2001). If these consequences of the thermocycling process in composite resins are observed, then it is reasonable to expect similar or inferior behavior from temporary resins.

In the present study, thermocycling simulated the intraoral condition, since the heating-cooling process results in repeated expansions and contractions of the material, (KAWANO et al., 2001) and ages the temporary resin. It can be inferred that the recurrent thermal variation promoted structural rearrangements and accelerated the oxidation process of tertiary amines. As a consequence, all temporary acrylic resins in this experiment showed color changes after 2,000 thermocycles. In a previous investigation, the color of an indirect composite resin was not influenced by the thermocycling 3,000 process, however, orange juice and coffee combined with thermocycling were able to discolor the polymeric material (ASSUNÇÃO e SOUZA et al., 2006). It is possible that thermal energy is sufficient to promote the BIS. - Decomposition of GMA leading to discoloration (ASSUNÇÃO e SOUZA et al., 2006). However, excessive exposure to water during the thermocycling process can negatively affect the mechanical properties of polymer-based materials, since the polymer is capable of absorbing water. In principle, the soluble components of the polymeric materials are removed and water is absorbed to replace the missing structures. If the material is kept immersed in water, the cross-link complex inflates and the strength of the polymer chain is reduced. It is assumed that water promotes hydrolysis at the matrix/filler interface and microcracks in the polymeric matrix, which contributes to the reduction of resin properties (ASSUNÇÃO e SOUZA et al., 2006).

In the literature, there is insufficient evidence correlating the number of thermal cycles and the clinical use of acrylic resins. However, it is estimated that 10,000 cycles would correspond to one year of clinical use of the provisional resin (CHAVES et al., 2009; MORRESI AL et al., 2014). Therefore, the 2,000 thermal cycles could be related to a few months of clinical use, which generally corresponds to a period of oral rehabilitation.

The second factor evaluated was the ability of an antiseptic mouthwash to stain the temporary resins tested. The use of chlorhexidine gluconate 0.12% during prosthetic rehabilitation is recommended to maintain gingival health and many authors attest to its efficiency (SILVA et al., 2007; MORRESI AL et al., 2014). On the other hand, this specific antimicrobial mouthwash can promote discoloration of the acquired film and coloration of materials. Polymer-based (CAL E et al.,



2007; MORRESI AL et al., 2014; GALE MS et al., 1999; CLAYDON N et al., 2001; LEE YK et al., 2006; TIEH MT et al., 2022).

After thermocycling and immersion in chlorhexidine (T3), significant discoloration was observed in the PMMA resins. (HIRAISHI N et al., 2008) observed that PMMA resins are more susceptible to staining with chlorhexidine, since these resins are highly hydrophilic. This observation could explain the behavior of PMMA. Despite this, although PMMA resins showed color changes statistically exacerbated by chlorhexidine, among the resins tested, Duralay® resin (PMMA) was the only resin that exhibited a color value (ΔE) greater than 3.3. The color grade expressed as ΔE indicates the color value of a given polymer material. ΔE value ranging from 2 to 3 is considered an acceptable color value, while ΔE greater than 3.3 is considered a clinically unacceptable value for resins. In a previous report, (CAL E et al., 2007) also noted a clinically noticeable staining ($\Delta E > 3.4$) of an acrylic resin exposed to mouthwashes such as chlorhexidine.

No comparisons were made between the tested resins, as commercially available provisional resins have different color scales and this fact excludes the possibility of an adequate statistical analysis (HASELTON DR et al., 2005).

The newest commercially available acrylic resins present innovative characteristics in their composition, such as the addition of glass powder, silica and microparticles that aim to develop a durable polymer-based provisional material (HIRAISHI N et al., 2008; SCHULZE KA et al., 2003; ALHOTAN A et al., 2022; ZAFAR MS et al., 2020; ALDEGHEISHEM A et al., 2021). Previously, SCHULZE KA et al., (2003) observed that resins with lower amounts of inorganic fillers (p.g.%) showed the highest levels of discoloration under accelerated aging. Additionally, (VICHI A et al., 2004), reported that the size and distribution of filler particles of composite resins directly correlate with the discoloration of composite resins subjected to aging in water. According to these authors, as the matrix-filler interface is one of the weakest points of the composite with high sensitivity to water sorption, it is likely that the hydrolytic degradation of this interface changes the way light is scattered by the particles.

To reverse the adverse effects promoted by chlorhexidine on polymer-based materials, recent mouthwashes containing polyvinylpyrrolidone (PVP) are available, which supposedly reduces coloring (MORRESI AL et al., 2014; RASZEWSKI Z et al., 2021). However, the discoloration of materials still appears as a negative factor. Effect, since the reduction of staining is only achieved with lower concentrations of chlorhexidine. On the other hand, decreasing the concentration of chlorhexidine may reduce its ability to inhibit biofilm growth and its effectiveness as an antiseptic mouthwash. The alternative hypothesis is



partially accepted, since thermal variation and immersion in chlorhexidine promoted color changes of PMMA resins.

Further in situ research understanding the effects of thermocycling and chlorhexidine on the color stability of acrylic resins is needed to validate the results of this in vitro experiment.

CONCLUSION

Thermocycling significantly influenced the color changes of the provisional acrylic resins tested. Chlorhexidine also affected the color stability of PMMA resins.

DISCLOSURE STATEMENT

There are no competing financial interests.

BIBLIOGRAPHIC REFERENCES

1. Christensen GJ. The fastest and best provisional restorations. J Am Dent Assoc. 2003 May 134(5): 637-639.
2. Haselton DR, Diaz-Arnold AM et al. Color stability of provisional crown and fixed partial denture. J Prosthet Dent. 2005 Jan 93(1):70-5.
3. Guler AU, Ylmaz F et al. Effects of different drinks on stainability of resin composite provisional restorative materials. J Prosthet Dent. 2005 Aug 94(2):70-5.
4. Rutkúnas V, Sabaliauskas V et al. Effects of different food colorants and polishing techniques on color stability of provisional prosthetic materials. D Materials J. 2010 Mar 29(2):167-176.
5. Alves PV, Lima Filho RM et al. Surface roughness of acrylic resins after different curing and polishing techniques. Angle Orthodontist. 2007 Jan 77(3):528-31.
6. Silva Filho CE, Martins da Silva EM. Avaliação da rugosidade superficial de resinas acrílicas submetidas à ciclagem térmica. Rev Fac Odontol Araçatuba. 2006 Jan-Jun (1) 27:28-33.



7. Zimmerli B, Koch T et al. The influence of toothbrushing and coffee staining on different composite surface coatings. Clin Oral Investig. 2011 Feb 18. [Epub ahead of print].
8. Silva WC, Silva DG et al. Effect of a mouthwash containing 0,12% of chlorhexidine on plaque inhibition and dental and tongue extrinsic staining. Rev Perionews. 2007 Jul (6): 229-34.
9. Cal E, Güneri P et al. Digital analysis of mouthrinses' staining characteristics on provisional acrylic resins. J Oral Rehabil. 2007 Apr;34(4):297-303.
10. Shinkai RS, Cury AA et al. In vitro evaluation of secondary caries development in enamel and root dentin around luted metallic restoration. Oper Dent. 2001 Jan-Fev 26(1):52-9.
11. Assunção e Souza RO, Michida SMA et al. Color stability of na indirect composite resin. Cienc Odontol Bras. 2006 Jul-Set 9(3):73-7.
12. Vichi A, Ferrari M et al. Color and opacity variations in three different resin-based composite products after water aging. Dent Mater. 2004 Jul 20(6):530-4.
13. Phoenix, RD. Denture base resins: Technical considerations and processing techniques, in Anusavice KJ: Phillips' Science of Dental Materials. Philadelphia, PA, Saunders. 1996 1(10):237-71.
14. [Turgut S](#) , [Bagis B](#) et al. Discoloration of provisional restorations after oral rinses. Int J Med Sci. 2013 Aug 30;10(11):1503-9.
15. Kawano F, Ohguri T et al. Influence of thermal cycles in water on flexural strength of laboratory-processed composite resin. J Oral Rehabil. 2001 Aug 28(8):703-7.
16. Chaves C de AL, Regis RR et al. Effect of ridge lap surface treatment and thermocycling on microtensile bond strength of acrylic teeth to denture base resins. Braz Dent J. 2009 20(2):127-31.
17. Morresi AL, [D'Amario M](#) et al. Thermal cycling for restorative materials: does a standardized protocol exist in laboratory testing? A literature review. J Mech Behav Biomed Mater. 2014 Jan; 29: 295-308.



18. Gale MS, Darvell BW. Thermal cycling procedures for laboratory testing of dental restorations. *J Dent.* 1999 Feb 27(2):89-99.
19. Claydon N, Addy M et al. Studies on the effect of polyvinyl pyrrolidone on the activity of chlorhexidine mouthrinses: plaque and stain. *J Clin Periodontol.* 2001 Jun 28(6):558-64.
20. Lee YK, Powers JM. Influence of salivary organic substances on the discoloration of esthetic dental materials – a review. *J Biomed Mater Res B Appl Biomater.* 2006 Feb 76(2):397-402.
21. Tieh MT, [Waddell](#) JN et al. Optical Properties and Color Stability of Denture Teeth-A Systematic Review. *J Prosthodont.* 2022 Jun;31(5):385-398.
22. Hiraishi N, Yiu CK et al. Chlorhexidine release and water sorption characteristics of chlorhexidine-incorporated hydrophobic/hydrophilic resins. *Dent Mater.* 2008 Oct 24(10):1391-9.
23. Schulze KA, Marshall SJ et al. Color stability and hardness in dental composites after accelerated aging. *Dent Mater.* 2003 Nov 19(7):612-9.
24. [Alhotan](#) A, [Elraggal](#) A et al. Effect of Different Solutions on the Colour Stability of Nanoparticles or Fibre Reinforced PMMA. *Polymers (Basel).* 2022 Apr 8;14(8):1521.
25. [Zafar](#) MS. Prosthodontic Applications of Polymethyl Methacrylate (PMMA): An Update. *Polymers (Basel).* 2020 Oct 8;12(10):2299.
26. [Aldegheishem](#) A, [AlDeeb](#) M et al. Influence of Reinforcing Agents on the Mechanical Properties of Denture Base Resin: A Systematic Review. *Polymers (Basel).* 2021 Sep 13;13(18):3083.
27. [Raszewski](#) Z, [Nowakowska](#) D et al. The Effect of Chlorhexidine Disinfectant Gels with Anti-Discoloration Systems on Color and Mechanical Properties of PMMA Resin for Dental Applications. *Polymers (Basel).* 2021 May 29;13(11):1800.