Asian Journal of Chemistry Vol. 22, No. 9 (2010), 6669-6676

Evaluation of Colour Changed of Acrylic Resin Materials in the Different Solutions

ZEYNEP YESIL DUYMUS*, NURAN YANIKOGLU and MUSTAFA ARIK†
Department of Prosthodontics, College of Dentistry, Atatürk University, Erzurum, Turkey
Fax: (90)(442)2360945; Tel: (90)(442)2311781; E-mail: zyesilz@hotmail.com

Colour stability of acrylic resin materials for denture base is important in their clinical performance. Present study evaluated the effect of material testing surface (rough and smooth), polymerization techniques (heat and auto-polymerized) and different solutions (coffee, tea, Turkish coffee, mouth wash and distilled water) on the colour stability of two acrylic resin materials. Thirty disks of each material (50 mm × 0.5 mm) were prepared. The specimens of each material were divided in two groups. The first groups were abraded with silicon carbide paper. The second groups were polished with pumice and white diamond. Twenty-four specimens were evaluated before and after 1, 7 and 30 days of immersion in different staining solutions and distilled water. Six specimens were evaluated before and after 6, 12 and 24 h of immersion in mouth wash. Colour changes were monitored by using the UV/vis/NIR spectrophotometer. As a results, material testing surface, staining solutions and immersion time were significant factors that affected the colour stability ($p < 0.001$). The auto-polymerized acrylic resin specimens showed high levels of colour changes in Turkish coffee. The heat polymerized acrylic resin materials were more colour stable than auto-polymerized acrylic resin specimens.

Key Words: Auto-polymerized acrylic resin material, Heat polymerized acrylic resin, Discolouration.

INTRODUCTION

The discolouration of resin restorations can result in an esthetic problem. Minimizing colour change is a factor that should be used in the selection of materials and techniques[1,2]. Discolouration may result from several factors. Impurities, incorporated during manufacturing or manipulation, can lead to colour changes. Most materials used for resin restorations are subject to sorption, a process of absorption and adsorption of liquids dependent upon environmental conditions. Technical variables that result in porosity or a surface quality conductive to the accumulation of debris may lead to significant discolouration[1,3]. For the best esthetic effect, the material should be translucent. Colour and translucency should be maintained during processing and these resins should not get stained or change colour in

†Department of Chemistry, College of Arts and Sciences, Atatürk University, Erzurum, Turkey.

clinical use. The colour stability criteria may provide important information on the serviceability of these materials[4].

Polymethyl methacrylate absorbs water slowly over time. Discolouring of the denture base polymers may be caused by the oxidation of the amine accelerator or by the penetration of coloured solutions[5,6]. Colour changes of denture base materials during exposure to oral fluids and denture cleaners have been reported[4-7]. There is evidence that beverages such as tea, coffee and wine significantly increase the development of stain on enamel and acrylic resin[6,8].

In addition, proprietary mouth washs are also added to these discolourizing factors[9,10], because their use has become popular recently[11]. Besides being an effective caries and gingivitis control method and topical relief measure in oral lesions, people tend to use mouth washs for social and cosmetic reasons[10-13].

Clinical studies have demonstrated that the denture base polymers tend to discolour during service in the oral environment[2,5,6,14,15]. Discolouration of the denture base polymers may be caused by intrinsic and extrinsic factors[5]. Extrinsic factors for discolouration include staining by adhesion or penetration of colourants as a result of contamination from exogenous sources, coffee and tea and coloured solutions[15-17].

Heat-polymerized acrylic materials have exhibited satisfactory colour stability, but auto-polymerized acrylic materials have shown relatively poor colour stability[18].

The purpose of this study *in vitro* was to evaluate the discolouration of heat and auto-polymerized acrylic denture base materials immersed in different solutions (tea, coffee, Turkish coffee, distilled water and mouth wash) at 37 ± 1 °C.

EXPERIMENTAL

Three staining solutions (coffee, tea and Turkish coffee), one mouth wash and distilled water were used and their effects on heat-polymerized (methylmethacrylate, QC-20, denture base material, Veined, DeTrey, Dentsply Caulk, Milford, DE 19963-0359, England) and auto-polymerized acrylic resin materials (RR Self-cured repair material, dark veined, DeTrey, Weybridge, surrey KT15 SE. England).

Thirty specimen disks, 50 mm diameter and thickness of 0.5 mm, were prepared from each material. The specimens were prepared as described in the American Dental Association's (ADA) specification No. 12[19] to evaluate colour changes mare accurately.

The heat-polymerized and auto-polymerized acrylic materials were processed according to the manufacturer's recommendations. All materials supplied in a powder and liquid form were mixed at a ratio of 7 mL of liquid to 10 g of powder. Half of the specimens were allowed to undergo short-time polymerization in a water bath at 72 °C for 1.5 h, following by 0.5 h boiling in 100 °C water. The auto-polymerized acrylic specimens were prepared in the same molds and remained at room temperature (22 ± 1 °C) for 0.5 h.

After polymerizing, the specimens of each material were divided into two groups. Fifteen discs were abraded on both sides with silicon carbide paper (No. 600, 3 m dental, Minn.) and remaining disks were polished for smooth surface. The disks were polished to a high gloss using fine powder of pumice and white diamond. Smooth and rough specimens were divided into 5 test groups for exposure to one of following conditions: Coffee, tea, Turkish coffee and distilled water at a constant temperature of 37 ± 1 °C for 1, 7 and 30 days. Mouth wash for 6, 12 and 24 h.

Each experimental group consisting of 6 specimens (rough and smooth) prepared from each test material was stored in 20 mL of one of the mouth wash (Andorex, Kim Pa, Istanbul, Turkey) for 6, 12 and 24 h. The period of 12 h was reported as the equivalent time to 1 year of 2 min daily mouth rinse use[9]. To the prepared coffee solution, 15 g of coffee (Nescafe classic, Nestlé, Société des Produits, S.P.N., Switzerland) was poured into 500 mL of boiling distilled water. The tea solutions were prepared by immersing 5 prefabricated doses of tea (Lipton, yellow label, Çorlu, Turkey) into 500 mL of boiling water for 10 min.

Turkish coffee was prepared from 5-7 g a commercial brand (Kurukahveci Mehmet Efendi, TS 3117, Y. Dudullu, Istanbul) in 65 mL distilled water. The powder was mixed with cold water and boiled for few minutes. Distilled water was used as a control solution. During this period the staining solutions were changed once a week.

Following the removal of the specimens from the staining solutions, they were dipped in distilled water and rinsed by moving them up and down 10 times. Excess fluid on the surfaces was removed using a tissue paper. After measurements of colour changes were carried out at time intervals indicated, the specimens were immersed in fresh staining solutions.

Colour changes of the specimens were determined through absorption measurements of the specimens by using a UV/vis/ NIR spectrophotometer (Shimadzu UV-3101 PC UV/vis/NIR scanning spectrophotometer). The instrument used for the measurement is a double beam spectrophotometer. The absorbance of each specimen was determined by using 500 nm monochromatic light. The measurements of colour changes of the specimens were made in three randomly selected areas near the center of each specimen. The average of the three readings was recorded and the mean of each material was calculated. The absorption measurements at the wavelength indicated were directly correlated with the colour of each sample before and after treatments with staining solutions, mouth wash and distilled water. Therefore absorbance of the specimens was determined by using Lambert-Beer Law given as[20]:

$$A = \varepsilon \cdot b \cdot C$$

where A = absorbance of specimen which is unitless, C = concentration of the acrylic material in g, ε = absorption coefficient that is 46.06 g^{-1} cm^{-1} determined at 500 nm for the specimen and b = thickness of specimen disc that is 0.5 mm. This formula was used for the determination of colour changes of the specimens. For

this purpose, the absorption of each disc was recorded by taking air as reference before the treatment with colour degrading materials. Then, the values of these absorption measurements were recorded. After this point, the specimens were treated with staining materials and mouth wash, then, absorption measurements were repeated under the same conditions. By taking absorbance differences of the specimens with treated colour degrading solutions and without treatments, we determined ΔA values that represent colour differences of each disc according to the beginning conditions. Therefore, ΔA values were used for the interpretation of experimental findings.

All data recording were taken by the same investigator to minimize inconsistency of technique. Statistical analysis was performed using ANOVA and Paired t-test.

RESULTS AND DISCUSSION

The ΔA means and standard deviations for all materials after stored in the staining solutions and distilled water are graphically displayed in Figs. 1 and 2. Results of the repeated measures ANOVA indicated that the effects of all factors and all possible interactions among them, were statistically significant ($p < 0.001$). Auto-polymerized acrylic material-Turkish coffee combinations revealed the highest ΔA value (1.12). Similar behaviour was observed with the coffee (ΔA = 0.98).

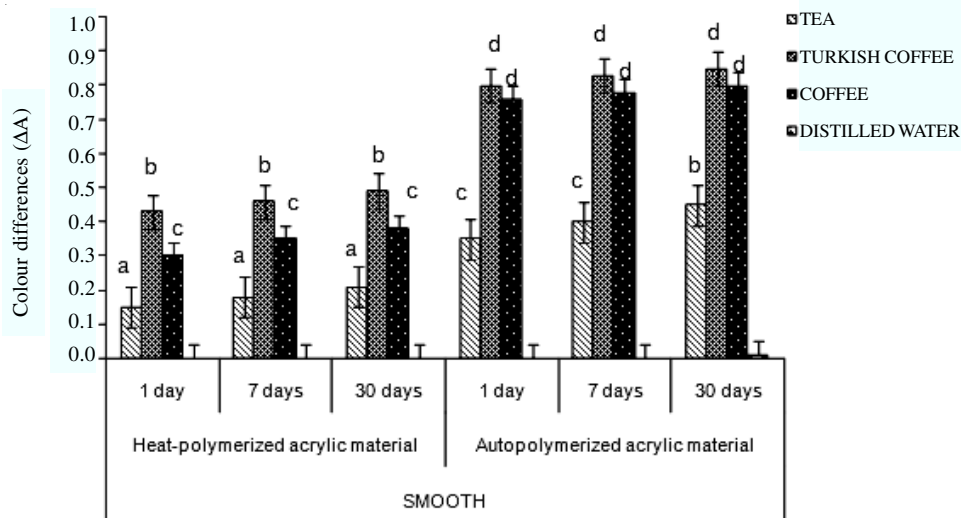

Fig. 1. Mean colour differences (ΔA) and standard deviations of smooth acrylic materials in different solutions. Identical letters indicate that the values are not statistically significant difference ($p < 0.001$)

After 30 days, the rough specimens darkened significantly more than the smooth discs ($p < 0.001$). Polymerization techniques had significant effect upon the amount

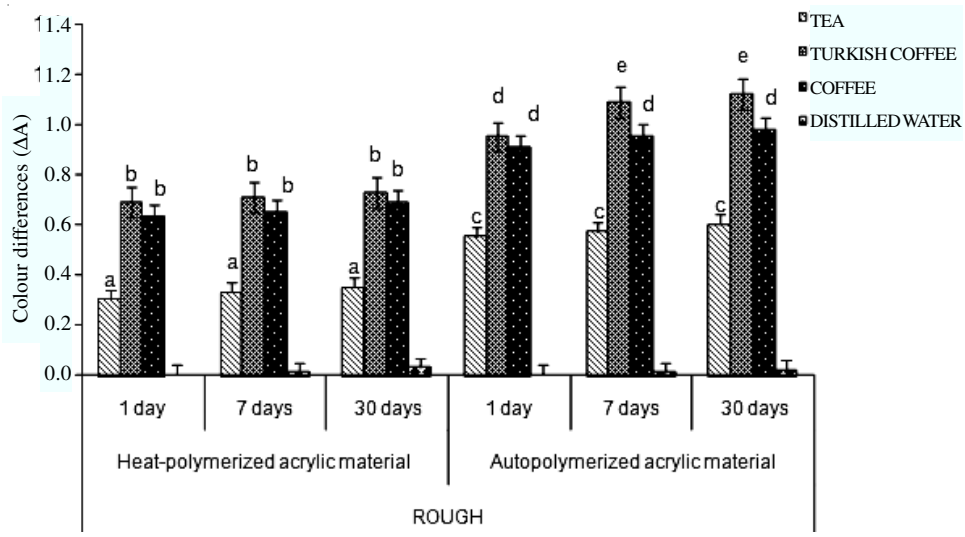

Fig. 2. Mean colour differences (ΔA) and standard deviations of rough acrylic materials in different solutions. Identical letters indicate that the values are not statistically significant difference ($p < 0.001$)

of colour change. The auto-polymerized acrylic material with rough surface was darkened significantly more than the other material. The material showing the least amount of colour change at 30 days was smooth and heat-polymerized acrylic material.

All the specimens displayed colour changes after immersion into the mouth rinse; however, the results showed that the effects of the mouth wash on the colour change of the materials were not different from that of distilled water ($\Delta A = 0.03 \pm 0.03$ in distilled water). The ΔA values in mouth wash after 24 h were below 0.09 given in Fig. 3.

Colour change of aesthetic materials has been reported to be dependent on the brand, the shade of the material and the finishing techniques[21-23]. The rough surfaces darkened much more than smooth specimens[1,24].

The variables used in this study were testing surface, curing technique and staining solution. The heat-polymerizing acrylic resin materials exhibited the least darkening, compared with auto-polymerizing acrylic materials. The stains were coffee and tea solutions and immersion time was up 30 days. Rough materials darkened significantly more than polished materials and curing technique have had effects on colour stability.

Scotti *et al.*[25] concluded that synthetic saliva and coffee produced the greatest darkening in restorative materials. Büyükyilmaz *et al.*[24] reported that denture base polymers were relatively colour stable when immersed in water at 50 ± 1 °C. The denture base materials were stained by coffee and tea solution.

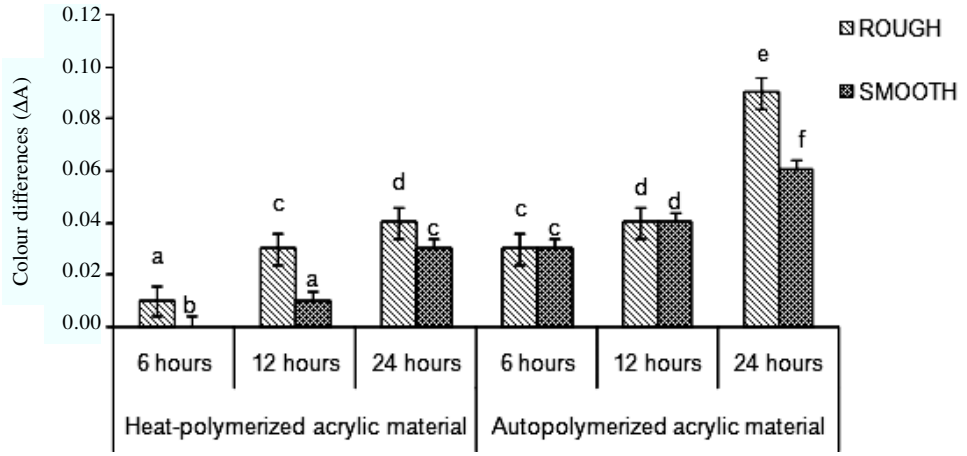

Fig. 3. Mean colour differences (ΔA) and standard deviations of acrylic materials in mouth wash. Identical letters indicate that the values are not statistically significant difference ($p < 0.001$)

Um and Ruyter[14] reported approximately twice as much discolouration in tea as in coffee for resin based veneering materials. Crispin and Caputo[1] reported that materials with rough surfaces darkened much more than smooth samples. The tea and coffee solution caused the greatest amount of darkening over a 1 month period.

Hersek *et al.*[26] have reported that the colour changes exhibited by all specimens after the 6th month were at clinically acceptable levels. The resin specimens absorbing these solutions used in colouring of food also may undergo staining. The absorption is undoubtedly due primarily to the polar properties of resin molecules.

After 30 days, rough and auto-polymerizing acrylic specimens were darkened significantly more than all other materials. After a 30 day immersion in coffee, all materials exhibited greater colour changes compared with tea, which is in agreement with the results of Yannikakis *et al.*[27] and Scotti *et al.*[25].

Time was found to be a critical factor for colour stability and staining solution of provisional restorative materials[15,27]. Koumjian *et al.*[8] found that acceptable colour changes of a short-term (5 weeks or less) period.

In the present study, paired t-tests showed that, as immersion time increased, colour changes became more intensive. All materials showed noticeable colour changes. These results are in agreement with the studies previously mentioned[1,8,14,25,27].

Koumjian *et al.*[8] evaluated the *in vivo* discolouration of 7 resins over a 9 week period. They found that heat-polymerized methylmethacrylate resin showed significantly less staining than the cold-cured methylmethacrylate resin.

In the present study, the rough auto-polymerized acrylic resin material at both 7 and 30 days had the greatest colour change with a ΔA of 1.09 and 1.12, respectively (in Turkish coffee). Heat-polymerized acrylic resin was the more colour stable. While colour changes may be attributed to excessive residual monomer, heat-

polimerized acrylic material ΔA of 0.73 was significantly less than auto-polymerized acrylic resin material at 30 days (in Turkish coffee).

The colour changes of the materials in the mouth wash after 12 h were not different from that of control solution (distilled water). These findings are in good agreement with the results of Gürgan *et al*.[9] and Gürdal *et al*.[28].

The immersion duration of the materials into the test solutions affects the colour changes[15]. In the present study, the specimens showed colour changes after 24 h of immersion in mouth wash. Colour change of denture base materials was also observed after the aging process[18] and immersion in disinfectants[29].

Conclusion

This study evaluated the colour stability of denture base materials when they were exposed to coffee, tea, mouth wash, Turkish coffee. Under the conditions of this study of following conclusions can be made: Staining solutions, polymerizing technique, surface treatments and immersion time are significant factors affecting colour stability. Turkish coffee solution exhibited more staining capacity than the other solution. Auto-polymerized acrylic resin showed the greatest ΔA values in Turkish coffee and coffee solutions. Rough materials darkened significantly more than polished materials. The smooth specimens of the heat-polymerized acrylic materials in mouth wash for 24 h were showed the least colour change but the colour change was not different from that in distilled water.

REFERENCES

1. B.J. Crispin and A.A. Caputo, *J. Prosthet. Dent*., **42**, 27 (1979).
2. T. Fusayama and T. Hirano, *J. Prosthet. Dent*., **25**, 532 (1971).
3. D.J. Pipko and M. El-Sadeek, *J. Dent. Res.*, **51**, 689 (1972).
4. J.L. Shotwell, M.E. Razzoog and A. Koran, *J. Prosthet. Dent.*, **68**, 836 (1992).
5. E. Asmussen, *Acta Odontol. Scand.*, **41**, 11 (1983).
6. W.D. Cook and M.P. Chang, *Biomaterials*, **6**, 257 (1985).
7. P. Yaman, M.E. Razzoog and H.E. Brandau, *J. Dent. Res.*, **67**, 213 (1988).
8. J.H. Koumjian, D.N. Firtell and A. Nimmo, *J. Prosthet. Dent.*, **65**, 740 (1991).
9. S. Gürgan, A. Öner and H. Köprülü, *J. Oral Rehabil.*, **24**, 244 (1997).
10. B. Penugonda, L. Settembrini, W. Scherer, E. Hittellman and H. Strassler, *J. Clin. Dent.*, **5**, 60 (1994).
11. E. Gagari and S. Kabani, *Oral Surg. Oral Med. Oral Pathol. Oral Radio. Endodon.*, **80**, 432 (1995).
12. D.M. Winn, *Am. J. Clin. Nutr*., **61**, 437S (1995).
13. D.M. Winn, W.J. Blot, J.K. McLaughlin, D.F. Austin, R.S. Greenberg, S. Preston-Martin, J.B. Schoenberg and J.F. Fraumeni, *Cancer Res.*, **51**, 3044 (1991).
14. C.M. Um and I.E. Ruyter, *Quintessence Int.*, **22**, 377 (1991).
15. Z.A. Khokhar, M.E. Razzoog and P. Yaman, *Quintessence Int.*, **22**, 733 (1991).
16. W.T. Wozniak, T.P. Muller, R. Silverman and J.B. Moser, *J. Oral Rehabil.*, **8**, 333 (1981).
17. C.N. Raptis, J.M. Powers, P.L. Fan and P.L. Yu, *J. Oral Rehabil.*, **9**, 367 (1982).
18. K.B. May, M.E. Razzoog, A. Koran and E. Robinson, *J. Prosthet Dent.*, **68**, 78 (1992).
19. Council on Dental Materials and Devices, Revised American Dental Association Specification No. 12 for Denture Base Polymers, *J. Am. Dent. Assoc*., **90**, 451 (1975).
20. G.D. Christian and J.E. O'Reilly, Instrumental Analysis, Allyn and Bacon Inc., Massachusetts, edn. 2 (1986).

21. O.F. Makinson, *Aust. Dent. J*., **34**, 154 (1989).
22. H. Uchida, J. Vaidyanathan, T. Viswanadhan and T.K. Vaidyanathan, *J. Prosthet Dent.*, **79**, 372 (1998).
23. W.W. Dodge, R.A. Dale, R.L. Cooley and E.S. Duke, *Dent. Mater*., **7**, 18 (1991).
24. S. Büyükyilmaz and I.E. Ruyter, *Int. J. Prosthodont*, **7**, 372 (1994).
25. R. Scotti, S.C. Mascellani and F. Forniti, *Int. J. Prosthodont*, **10**, 164 (1997).
26. N. Hersek, S. Canay, G. Uzun and F. Yildiz, *J. Prosthet Dent.*, **81**, 375 (1999).
27. S. Yannikakis, A.J. Zissis, G.L. Polyzois and C. Caroni, *J. Prosthet Dent*., **80**, 533 (1998).
28. P. Gürdal, B.G. Akdeniz and B.H. Sen, *J. Oral Rehabil*, **29**, 895 (2002).
29. M. Tsun, G.H. Johnson and G.E. Gordon, *J. Prosthet Dent*., **77**, 197 (1997).

(*Received*: 29 August 2009; *Accepted*: 31 May 2010) AJC-8748

ERRATUM

Asian Journal of Chemistry Vol. 22, No. 3 (2010), 1689-1692

Synthesis and Crystal Structure of 1-(2-(1*H*-Benzoimidazol-1-yl)ethyl)-1*H*-benzoimidazol-3-ium Chloride Hydrate

JUN YANG, CAI FENG BI*, TINGTING YOU† and YUHUA FAN*
College of Chemistry and Chemical Engineering,
Ocean University of China, Qingdao-266100, P.R. China
Fax: (86)(532)84022750; Tel: (86)(532)84022946; E-mail: qustchemistry@126.com

1. In ACKNOWLEDGEMENTS

Kindly read (**Grant No. 20971115**) not as (Grant No. 0971115).