

# Color Stability of Esthetic Ion-releasing Restorative Materials Subjected to pH Variations

*José Carlos Pettorossi Imparato, DDS, MS, PhD    Anselmo Garcia, DDS  
Clarissa Calil Bonifácio, DDS    Lisa Scheidt, DDS, MS  
Daniela Prócida Raggio, DDS, MS, PhD  
Fausto Medeiros Mendes, DDS, MS, PhD  
Mário Vedovello Filho, DDS, MS, PhD*

## ABSTRACT

**Purpose:** The purpose of this study was to evaluate the color stability of 3 types of fluoride-releasing restorative esthetic materials when exposed to pH variations: (1) a polyacid-modified resin composite; (2) a resin-modified glass ionomer cement; and (3) a conventional glass ionomer cement in 2 different storage conditions.

**Methods:** Five samples of each material were stored in artificial saliva (pH 6.8) for 28 days. Five other samples were submitted for a pH-cycling during the same 28 days, which were immersed daily in a demineralizing solution (pH 4.5) for 7.5 hours and in a remineralizing solution (pH 7.0) for 16 hours. Color parameters were analyzed in a reflectance spectrophotometer to evaluate the color changes using the CIE L\*a\*b\* measuring system, after 7 and 28 days. The results were submitted to analysis of variance and Tukey tests.

**Results:** There was no difference in the same material concerning the different storage conditions. Conventional glass ionomer cement, however, showed greater color stability in relation to resin-modified glass ionomer cement ( $P < .05$ ) and to polyacid-modified resin composite ( $P < .01$ ).

**Conclusions:** There were no significant color changes due to pH variations, and conventional glass ionomer cement presented greater color stability. (J Dent Child 2007;74:189-93)

KEYWORDS: COLOR STABILITY, RESTORATIVE MATERIALS, GLASS IONOMER CEMENTS

*Dr. Imparato is assistant professor, Department of Pediatric Dentistry, School of Dentistry, University of São Paulo, São Paulo, Brazil, Post-graduate professor, Department of Pediatric Dentistry, School of Dentistry of São Leopoldo Mandic, Campinas, Brazil, and responsible for Pediatric Dentistry Department of Uniararas, Araras, Brazil; Dr. Garcia has a private practice in São Paulo, Brazil; Dr. Bonifácio is graduate student at Department of Pediatric Dentistry, School of Dentistry, University of São Paulo, São Paulo, Brazil; Dr. Scheidt is graduate student, São Leopoldo Mandic, Campinas, Brazil; Dr. Raggio is professor at Uniararas, Araras, Brazil; Dr. Mendes is assistant professor, Department of Pediatric Dentistry, School of Dentistry, University of São Paulo, São Paulo, Brazil; and Dr. Vedovello is professor at Uniararas, Araras, Brazil. Correspond with Dr. Imparato at jcpimparato@yahoo.com.br*

Current restorative techniques have been introduced to improve esthetic results. A great number of in vitro studies have been performed using different restorative materials in order to achieve long-term stability. Spectrophotometry and colorimetry are used to evaluate several parameters related to color stability.<sup>1-5</sup>

Currently, fluoride-releasing esthetic restorative materials have been extensively used due to their preventive effect on dental caries.<sup>6-10</sup> The color stability of these restorative materials has been a challenge to dentistry,<sup>9,11-14</sup> once the exposure to intrinsic and extrinsic factors of oral cavity can determine the color change of restorations.<sup>15-17</sup> Furthermore, children with poor oral hygiene could be more susceptible to the staining around marginal restorations because of caries lesions and hypomineralized dental structures.<sup>11,13</sup>

The conventional glass ionomer cement (GIC) is a fluoride-containing restorative material with nonsatisfactory color stability.<sup>9,11,13,18</sup> Different restorative materials—such as high-viscosity GIC, polyacid-modified resin composite (PMRC), or compomers and resin-modified GIC—have been developed to improve some physical properties of the materials, including the color stability.<sup>9,10,19-21</sup> A higher level of released fluoride has been observed from GIC samples stored in acid conditions than those stored in neutral or basic solutions.<sup>21-23</sup> The greater ion exchange could produce more pronounced color changes in the material exposed to constant pH changes.

Thus, the aim of the present study was to evaluate the color stability of 3 types of ion-releasing esthetic restorative materials immersed in a soft drink when exposed to pH variations.

## METHODS

Three esthetic, restorative, ion-releasing materials were used: a PMRC, a resin-modified GIC, and a conventional GIC with a high powder/liquid ratio indicated to atraumatic restorative treatment (ART; Table 1).

**Table 1. Restorative Materials, Brands, Manufacturers, and Color Shades Used in the Study**

Restorative material	Product name	Manufacturer	Shade
Resin-modified glass ionomer	Vitremer <sup>a</sup>	3M/ESPE, St. Paul, Minn	Pedo
Conventional glass ionomer	Fuji IX <sup>b</sup>	GC America Co, Tokyo, Japan	U
Polyacid-modified resin composite	Compoglass <sup>c</sup>	Ivoclar Vivadent, Schaan, Liechtenstein	B1

The tooth-colored materials' chosen shades are usually used to restore primary teeth.

Thirty rectangle-shaped samples of each material (7 mm long, 4 mm wide, and 1.5 mm thick) were prepared. The resin-modified GIC and the conventional GIC were manipulated according to manufacturer's instructions and inserted into molds in a single insertion with a Centrix (DFL Ind. Com., RJ, Brazil) syringe. The PMRC was inserted in 3 increments. The resin-modified GIC and PMRC specimens were light cured for 40 seconds using a light-curing unit with an average light output of 720 mW/cm<sup>2</sup> (XL 1500, 3M/ESPE, St. Paul, Minn). The samples were incubated at 100% relative humidity at 37°C for 24 hours. Afterwards, the samples were polished with polishing disks (Soflex, 3M/ESPE).

After the polishing, the samples were rinsed and dried with paper tissue and the baseline color measurements were performed. Color evaluations were made in a reflectance spectrophotometer (Cintra 10 GBCUV, GBC Scientific Equipment, Dandenong, Australia), with color parameters based on average daylight (D65: 6504 K). The system's optical geometry consisted of a 45° illumination angle and

an observation angle of 2°. Calibration was made using a white standard.

The samples of each material were randomly allocated into 2 groups (N=5). In the first group, samples were stored in artificial saliva. The composition of the artificial saliva was: (a) 0.3 mg CaCl<sub>2</sub>; (b) 0.004mg Na<sub>2</sub>HPO<sub>4</sub>; (c) 0.5 mg urea; (d) 0.0008 mg Na<sub>2</sub>S; (e) 0.0008 mg Na<sub>4</sub>P<sub>2</sub>O<sub>7</sub>; and (f) 0.2mg of NaCl for 500 ml of H<sub>2</sub>O adjusted to pH 7.0.<sup>24</sup>

The other samples were submitted for a pH-cycling. The samples were immersed daily in demineralizing solution for 7.5 hours and in a remineralizing solution for 16 hours. The demineralizing solution contained: (a) 2.2 mM CaCl<sub>2</sub>; (b) 2.2 mM NaH<sub>2</sub>PO<sub>4</sub>; and (c) 50 mM acetic acid adjusted to pH 4.8. The remineralizing solution contained: (a) 1.5 mM CaCl<sub>2</sub>; (b) 0.9 mM NaH<sub>2</sub>PO<sub>4</sub>; and (c) 0.15 M KCl adjusted to pH 7.0.

Both sample groups were removed daily from the storing solutions and immersed in a soft drink (Coca Cola, The Coca Cola Company, Atlanta, Ga) for 30 minutes. The experiment was performed for 28 days. New color evaluations were carried out after 7 and 28 days. Previous studies used cola to simulate the staining of restorative materials.<sup>8,14</sup>

The color stability was evaluated by determining the color variation ( $\Delta E^*$ ) between the final (f) and reference baseline (o) using the color measuring system CIE L\*a\*b\*. The average value ( $\Delta E^*$ ) was calculated for each group. The color changes were calculated by using the formula:

$$\Delta E^* = [(L_o^* - L_f^*)^2 + (a_o^* - a_f^*)^2 + (b_o^* - b_f^*)^2]^{1/2}$$

L\* is the reflection factor of luminescence, +a\* is red, -a\* is green, +b\* is yellow, and -b\* is blue.

The mean  $\Delta E^*$  values of the restorative materials were first analyzed for normality using the Kolmogorov-Smirnov test. Then, the mean color changes for the three restorative materials—stored in artificial saliva or submitted for pH-cycling after 7 and 28 days—were analyzed using a 3-way ANOVA with repeated measures. Mean pairs of mean were compared using the Tukey test. The statistical level of significance for all tests was  $P < .05$ .

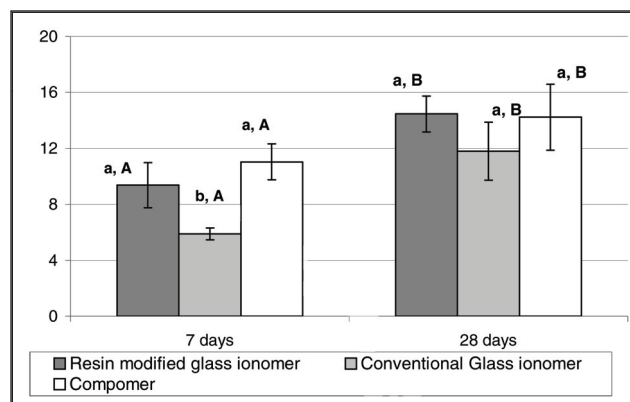
## RESULTS

When the color changes were analyzed according to the storage solution (independently of restorative material or period of storage), there was no statistically significant difference between  $\Delta E^*$  in the samples stored in artificial saliva (mean $\pm$ SD=11.13 $\pm$ 3.33) and in the de/remineralizing cycling (11.49 $\pm$ 4.16).

Comparing only the materials, however, conventional GIC presented significantly lower color change values (8.46 $\pm$ 3.18) than the resin-modified GIC (11.79 $\pm$ 3.34). Besides, resin-modified GIC presented lower changes than the PMRC (13.67 $\pm$ 2.75). Regarding the storage time, the samples stored for 28 days presented significantly higher color changes statistically (13.82 $\pm$ 2.82) than the samples stored for 7 days (8.80 $\pm$ 2.73).

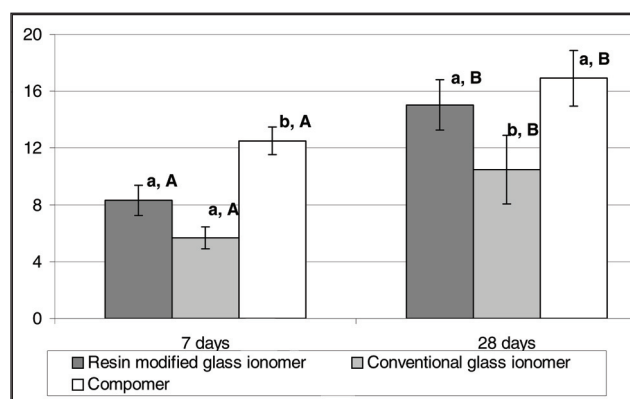
As there was no significant influence between storage solutions, the data from the samples stored in artificial saliva and submitted for pH-cycling were presented separately. In the samples stored in artificial saliva, the color changes

mean after 28 days was significantly higher statistically than that recorded after 7 days for all the materials. After 7 days, there were significantly lower color changes among conventional GIC compared with the resin-modified GIC ( $P<.05$ ) and with the PMRC ( $P<.01$ ; Figure 1). After 28 days, there was no significant change observed among the materials (Fig. 1).



**Figure 1.** Mean of color changes ( $\Delta E$ ) of different materials stored in artificial saliva measured after 7 and 28 days. Different lowercase letters indicate a statistically significant difference ( $P<.05$ ) among different restorative materials evaluated during the same period. Different uppercase letters indicate a statistically significant difference ( $P<.05$ ) between the same material evaluated during different periods. Vertical lines indicate the standard deviations.

Considering the samples submitted for pH-cycling, conventional GIC and resin-modified GIC presented lower color changes than PMRC after 7 days ( $P<.01$ ; Figure 2). After 28 days, significant color changes occurred in PMRC and in resin-modified GIC when compared with conventional GIC ( $P<.001$ ; Figure 2).



**Figure 2.** Mean of color changes ( $\Delta E$ ) of different materials stored in de/remineralizing cycling measured after 7 and 28 days. Different lowercase letters indicate a statistically significant difference ( $P<.05$ ) among different restorative materials evaluated during the same period. Different uppercase letters indicate a statistically significant difference ( $P<.05$ ) between the same material evaluated in different periods. Vertical lines indicate the standard deviations.

## DISCUSSION

Color stability in esthetic restorative materials represents a challenge in dentistry if the color change is still one of the major causes of restoration replacements in the labial surfaces of teeth.<sup>25</sup> Therefore, a great number of studies have been carried out using different restorative materials. Spectrophotometry and colorimetry have been the main tools to evaluate parameters related to color stability.<sup>1-5</sup>

Previous studies have shown that fluoride-releasing materials have a greater ion release when submitted to pH variations than when stored in artificial saliva or saline.<sup>21-23</sup> These facts could lead to lower color stability due to the great amount of ionic changes between the materials and the environmental solutions. A long-term study showed:

1. more fluoride being released from a conventional GIC in artificial saliva than a resin-modified GIC; and
2. the color stability of conventional GIC was worse than for the resin-modified GIC.<sup>7</sup>

GIC's compressive strength was inversely correlated with the amount of fluoride released.<sup>26</sup> Furthermore, some authors observed that GIC and PMRC samples stored in an acetic acid solution presented significant color changes.<sup>18</sup> Thus, the purpose of this study was to investigate the color stability of different ion-releasing restorative materials when exposed to pH variations.

Usually, fluoride-releasing materials (hydrophilic) are more susceptible to staining than hydrophobic materials (such as composite resins).<sup>9,27</sup> Nevertheless, fluoride-releasing restorative materials are the first choice when treating children with caries activity because GIC restorations have presented a lower incidence of secondary caries than amalgam.<sup>28</sup> Therefore, color stability of the fluoride-releasing materials in conditions of pH variation would be desirable. In this study, there was no difference between the materials immersed in artificial saliva and the materials submitted for pH-cycling. Based on this fact, the color stability of ion-releasing restorative materials would not be a great concern for patients with high dental caries activity.

This study's findings, however, should be interpreted in the context of study limitations. The present study was performed over 28 days. Longer periods could lead to more significant color changes, since the pH changes are shorter but more frequent in clinical conditions. Further studies with greater storage periods are necessary. Another factor is the immersion time in the staining solution. While most studies immerse the samples for longer periods in the dyes,<sup>5,8,9,14</sup> this study immersed the samples in a soft drink for 30 minutes daily. It can be argued that this period better simulates the time in which the restorations are exposed to pigmentation in the oral cavity. Another point concerns this study's in vitro design. Thus, further in vivo studies should be performed to test the color stability of the fluoride-releasing restorative materials exposed to different challenges in the oral cavity.

Previous studies have observed a greater color change in GICs and PMRCs.<sup>9,27</sup> This fact could be explained by the microfractures, which allow for stain penetration and material pigmentation.<sup>9</sup> Another study observed greater color change among PMRCs and resin-modified GICs compared with composites, but the conventional GIC was not used.<sup>8</sup> The PMRC was susceptible to staining with: (1) cola; (2) red wine; and (3) coffee staining.<sup>14</sup> In the present study, however, the conventional GIC presented greater color stability than the other tested materials.

This difference could be due to the fact that Fuji IX is a high-viscosity, studies using restorations with high-viscosity GIC have produced better clinical results than those with low-viscosity GIC.<sup>29</sup> Nevertheless, Fuji IX presented the worst color stability in a previous study.<sup>9</sup> In this latter study, however, more significant changes of this material occurred after ultraviolet light exposure. When the FUJI IX samples were immersed in a staining solution with no ultraviolet light exposure, there was no significant color changes compared with other materials.<sup>9</sup> Based on these facts, it seems that this material's color stability is more susceptible to ultraviolet light exposure than to staining with dyes.

Concerning the other materials, results obtained in the present study showed a greater color change of the PMRC and resin-modified GIC. Previous studies corroborate these results.<sup>8,9,12,14,18</sup>

## CONCLUSIONS

Based on this study's results, it can be concluded that the pH variations do not increase the color changes of fluoride-releasing dental materials. Conventional glass ionomer cement presents greater color stability to the dye staining.

## ACKNOWLEDGMENTS

The authors are very grateful to Drs. Paula Bartok and Rogério Sanchez (Faculdade de Odontologia da Universidade de São Paulo, São Paulo, Brazil) for their English corrections. The authors also wish to thank the Department of Operative and Restorative Dentistry of the School of Dentistry, University of São Paulo, São Paulo, Brazil, for technical support. The authors certify that they have no affiliation with or financial involvement in any organization or entity with a direct financial or personal interest in the subject matter or materials discussed in this manuscript.

## REFERENCES

1. Asmussen E, Hansen EK. Surface discoloration of restorative resins in relation to surface softening and oral hygiene. *Scand J Dent Res* 1986;94:174-7.
2. Klemetti E, Matela AM, Haag P, Kononen M. Shade selection performed by novice dental professionals and colorimeter. *J Oral Rehabil* 2006;33:31-5.
3. Ferracane JL, Moser JB, Greener EH. Ultraviolet light-induced yellowing of dental restorative resins. *J Prosthet Dent* 1985;54:483-7.
4. Satou N, Khan AM, Matsumae I, Satou J, Shintani H. In vitro color change of composite-based resins. *Dent Mater* 1989;5:384-7.
5. Um CM, Ruyter IE. Staining of resin-based veneering materials with coffee and tea. *Quintessence Int* 1991;22:377-86.
6. el Mallakh BF, Sarkar NK. Fluoride release from glass-ionomer cements in de-ionized water and artificial saliva. *Dent Mater* 1990;6:118-22.
7. Williams JA, Billington RW, Pearson GJ. A long-term study of fluoride release from metal-containing conventional and resin-modified glass ionomer cements. *J Oral Rehabil* 2001;28:41-7.
8. Abu-Bakr N, Han L, Okamoto A, Iwaku M. Color stability of compomer after immersion in various media. *J Esthet Dent* 2000;12:258-63.
9. Iazzetti G, Burgess JO, Gardiner D, Ripps A. Color stability of fluoride-containing restorative materials. *Oper Dent* 2000;25:520-5.
10. Garcia-Godoy F. Resin-based composites and compomers in primary molars. *Dent Clin North Am* 2000;44:541-70.
11. Hino DM, Mendes FM, de Figueiredo JL, Gomide KL, Imparato JC. Effects of plaque disclosing agents on esthetic restorative materials used in pediatric dentistry. *J Clin Pediatr Dent* 2005;29:143-6.
12. Chinelatti MA, Ramos RP, Chimello DT, Palma-Dibb RG. Clinical performance of a resin-modified glass-ionomer and two polyacid-modified resin composites in cervical lesions restorations: 1-year follow-up. *J Oral Rehabil* 2004;31:251-7.
13. Debner T, Warren DP, Powers JM. Effects of fluoride varnish on color of esthetic restorative material. *J Esthet Dent* 2000;12:160-3.
14. Fay RM, Walker CS, Powers JM. Discoloration of a compomer by stains. *J Gt Houst Dent Soc* 1998;69:12-3.
15. Powers JM, Fan PL, Raptis CN. Color stability of new composite restorative materials under accelerated aging. *J Dent Res* 1980;59:2071-4.
16. Ruyter IE, Nilner K, Moller B. Color stability of dental composite resin materials for crown and bridge veneers. *Dent Mater* 1987;3:246-51.
17. Dietschi D, Campanile G, Holz J, Meyer JM. Comparison of the color stability of 10 new-generation composites: An in vitro study. *Dent Mater* 1994;10:353-62.
18. Lim BS, Moon HJ, Baek KW, Hahn SH, Kim CW. Color stability of glass-ionomers and polyacid-modified resin-based composites in various environmental solutions. *Am J Dent* 2001;14:241-6.
19. Sidhu SK, Watson TF. Resin-modified glass ionomer materials. A status report for the American Journal of Dentistry. *Am J Dent* 1995;8:59-67.



20. McCabe JF. Resin-modified glass-ionomers. *Biomaterials* 1998;19:521-7.
21. Vieira AR, de Souza IP, Modesto A. Fluoride uptake and release by composites and glass ionomers in a high caries challenge situation. *Am J Dent* 1999;12:14-8.
22. Forss H. Release of fluoride and other elements from light-cured glass ionomers in neutral and acidic conditions. *J Dent Res* 1993;72:1257-62.
23. Hayacibara MF, Ambrozano GM, Cury JA. Simultaneous release of fluoride and aluminum from dental materials in various immersion media. *Oper Dent* 2004;29:16-22.
24. Osinaga PW, Grande RH, Ballester RY, Simionato MR, Delgado Rodrigues CR, Muench A. Zinc sulfate addition to glass-ionomer-based cements: Influence on physical and antibacterial properties, zinc, and fluoride release. *Dent Mater* 2003;19:212-7.
25. Kroeze HJ, Plasschaert AJ, van 't Hof MA, Truin GJ. Prevalence and need for replacement of amalgam and composite restorations in Dutch adults. *J Dent Res* 1990;69:1270-4.
26. Xu X, Burgess JO. Compressive strength, fluoride release and recharge of fluoride-releasing materials. *Biomaterials* 2003;24:2451-61.
27. Douglas WH, Craig RG. Resistance to extrinsic strains by hydrophobic composite resin systems. *J Dent Res* 1982;61:41-3.
28. Qvist V, Laurberg L, Poulsen A, Teglers PT. Eight-year study on conventional glass ionomer and amalgam restorations in primary teeth. *Acta Odontol Scand* 2004;62:37-45.
29. Frencken JE, Van 't Hof MA, Van Amerongen WE, Holmgren CJ. Effectiveness of single-surface ART restorations in the permanent dentition: A meta-analysis. *J Dent Res* 2004;83:120-3.