

Color Stability of Resin Cements after Accelerated Artificial Aging

Research Article

Running title: Color Stability of Resin Cements

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Abstract

Purpose: This investigation evaluated the intrinsic color stability of four resin cements after water storage and exposure to an artificial sunlight source in bulk and under a simulated ceramic restoration.

Materials and Methods: Cylindrical bulk cement specimens (n=8) measuring 8mm in diameter and 3mm in depth of G-CEM one (G), Rely-X Universal (R), Panavia V5 (P) and Variolink Esthetic (V) were prepared in a custom mold, visible light cured for 40 seconds and polished to a highly glossy surface. Ceramic specimens, 1 ± 0.1 mm in thickness X 12.8mm X 14.8mm wafers were cut from of GC Initial LiSi ceramic blocks and one side polished to a 4000 grit surface with abrasive papers. The wafers were placed in a custom mold with a 0.12 mm thick space for the resin cement and the cement placed and cured for 40 seconds through the ceramic. After water storage for 24 hours at 37° C baseline color measurements were made with the Vita EasyShade V instrument calculating Commission international de l'éclairage $L^*a^*b^*$ (CIELAB) values for each specimen. Each specimen was subjected to 16 four-hour aging cycles using a Xenon light source with an intensity of 0.4 W/m². 8 specimens of each cement were used as control with no accelerated aging. Delta E was measured at baseline, after 7 days and after the completed aging protocol. Two-way ANOVA (cement and measurement period) and post-hoc LSD test was used for determining differences among the cements.

Results: There were significant differences of delta E among the cements tested (p<0.05). R generated a delta E change greater than 3 in the bulk form.

Conclusion: There were significant differences in the measured delta E values for the cements exposed to accelerated aging.

Keywords: Ceramics; Color; Dental materials; Light curing; Resin cements

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Introduction

Advances in dental ceramics have made them a preferred choice for indirect dental restorations including veneers, inlays onlays and crowns. The natural appearing color and translucency of ceramics has made them uniquely suited for anterior restorations [1]. Clinical techniques have taken advantage of stronger ceramics in order to generate more conservative preparations leading to restorations with greater translucency [2,3]. Thus, the underlying tooth structure as well as the luting cement are critical influences on the shade matching of these restorations [4,5]. Therefore for long-term success, particularly of anterior restorations the color stability of the underlying luting cements is critical [6]. Resin-based cements are frequently employed for the placement of ceramic restorations because of

their numerous favorable properties. These include: low solubility, high mechanical properties, the ability to achieve a high bond to tooth structure and their ability to provide support for the intaglio surface of the dental ceramic [1]. However resin cements can undergo color change and degradation due to both extrinsic and intrinsic factors [7]. Intrinsic factors affecting discoloration are most especially related to the chemistry of the cement's constituent components. In particular the chemical activated self-cure and dual-cure cement systems may experience color change due to the oxidation of the reactive groups in the amine accelerators or inhibitors in the cement. Chemical decomposition of stabilizers and reactant chemicals often cause a change in the hue of cured cements often towards more yellow [8]. In addition color change can occur due to unreacted monomers [9] as well as

a result of physicochemical reactions resulting from UV radiation and water sorption [10].

Numerous investigations have been conducted to evaluate the color stability of resin cements subjected to a variety of accelerated aging conditions [11-16]. While useful in providing a basic understanding of the behavior of these cements, most investigations focused on the resin material itself and not the possible effects of the show-through of discoloration on a ceramic backed by the resin cement. This investigation was designed with two objectives: 1- to evaluate the intrinsic color stability of four resin cements after water storage and exposure to an artificial sunlight source and 2- to evaluate the color changes of a ceramic restorative material following placement of the resin cements.

Materials and Methods

Four dual-cured resin-based cements were evaluated in this investigation (Table 1). Cylindrical bulk cement specimens (n=8) (Figure 1) measuring 8mm in diameter and 3mm in depth were prepared in a custom mold, visible light cured for 40 seconds using A VALO curing light (Ultradent Products Inc., South Jordan UT USA) and polished to a highly glossy surface using the Sof-lex system (3M Oral Care, St. Paul MN, USA). Ceramic wafers 1 ± 0.1 mm in thickness wafers were cut from lithium disilicate ceramic blocks (GC Initial, LiSo Block, GC Tokyo, Japan) shade A1 and one side polished to a 4000-grit surface with abrasive papers (Figures 2,3 and 4). The non-polished surfaces of the ceramic wafers were treated with Monobond Etch and Prime (Ivoclar, Schaan, Liechtenstein). Using a custom mold with a cement space widow 0.12 mm in thickness (Figure 5), each cement (n=8) was placed in the mold (Figure 6) and the ceramic positioned over the cement with any excess cement carefully removed. Each specimen was light polymerized for 40 seconds through the ceramic wafer and the cement/ceramic assembly was removed from the mold (Figure 7).

In addition to the specimens for accelerated aging, eight bulk and eight cement wafer specimens were prepared as controls for each cement. The bulk cement specimens and cemented wafer assemblies for each cement were placed in water storage for 24 hours at 37° C. Following the initial storage period, baseline color measurements were made using the VITA Easshade V (Vita



Figure 1: Bulk cement specimen 8mm X 3 mm.



Figure 2: Initial LiSi lithium disilicate block used for ceramic wafer specimens.



Figure 3: Preparing 1mm thick wafer from ceramic block using a diamond saw.



Figure 4: Ceramic wafer 12.8 X 14.8 X 1mm used.

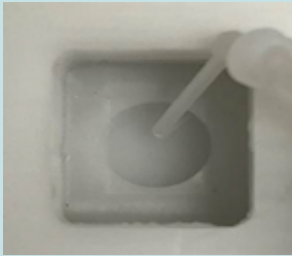


Figure 5: Custom mold with 0.12 mm well for cement for creating cement/wafer specimen.

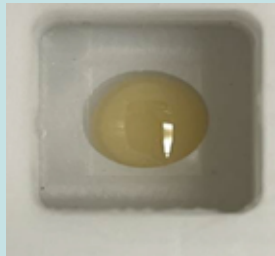


Figure 6: Cement placed in mold prior to placement of ceramic wafer.



Figure 7: Final cement/wafer assembly following visible light polymerization.



Figure 8: VITA Easyshade V device for measuring CIELAB $L^*a^*b^*$ values.

North America, Yorba Linda CA, USA.) using a standard white background. Measurements were made under standard lighting conditions D65, and the device was calibrated according to the manufacturer’s recommendations before each measurement. Three measurements were made and for each sample and the mean $L^*a^*b^*$ values were recorded for each specimen and each cement group [17].

Simulated accelerated aging was done using a xenon light source chamber (A-Sun Benchtop Xenon Light Accelerated Weathering Chamber, Sanwood Technology Co., Ltd, Guangdong,

Table 1: Resin Cements Used in this Study.

The material composition is based on the information published by the manufacturer.

Cement	Shade	Composition	Lot Number
G-CEM One (GC, Tokuo Japan) (G)	A2 Transluscent	fluoroaluminosilicate glass, UDMA, dimethacrylate, initiators, stabilizers, pigments, silicon dioxide, MDP, trimethoxysilane, 2-hydroxy-1,3-dimethacryloxypropane, 6-tert-butyl-2,4-xyleneol, 2,6-di-tert-butyl-p-cresol, EDTA disodium salt dehydrate, vanadyl acetylacetonate, TPO, ascorbic acid, camphorquinone, MgO	2210141 (2024-10-13) 2209291 (2024-09-28)
Panavia V5 (P) (Kuraray Noritake, Tokyo, Japan) (P)	A2	Bis-GMA, TEGDMA, Hydrophobic aromatic dimethacrylate, hydrophilic aliphatic dimethacrylates, Initiators, Accelerators, Silanated barium glass filler, Silanated fluoro- aluminosilicate glass filler, Colloidal silica, Silanated aluminium oxide filler, dl Camphorquinone, Pigments	BH0220 (2025-09-30)
RelyX Universal (3M Oral Care, St. Paul MN, USA) (R)	TR	phosphorus oxide, silane, terimethoxyctyl-hydrolysis product with silica, t-Amyl hydroxiperoxide, n2,6-di-tert-butyl-p-cresol, 2-HEMA, methyl methacrylate, acetic acid, copper slat, monohydrate diurethanedimethacrylate, ytterbium fluoride, 2-methyl-3(trimethoxysilyl)propyl ester and phenyltrimethoxysilane, TEGDMA, L-ascorbic acid, 6-hexadecanoate hydrate, titanium dioxide, triphenyl phosphite	9495612 (2024-04-03)
Variolink Esthetic (Ivoclar, Schaan, Liechtenstein) (V)	Light	UMDA, methacrylate monomers as 1,10-decandiol dimethacrylate, a-dimethylbenzyl hydroperoxide, initiators, stabilizers, pigments and inorganic fillers of ytterbium trifluoride, spheroid mixed oxide	Z049W4 (2025-03-15)

China.) Each specimen was subjected to 16 four-hour aging cycles at a wavelength range of 240 nm to 800 nm from the xenon source with an intensity of 0.4 W/m². Between each four-hour exposure, the specimens were stored in water at 37° C. Before each accelerated aging cycle, the specimens were removed from water and allowed to air dry before being placed in the xenon chamber. Specimens were also air dried before making color measurements after removal from water. CIELAB $L^*a^*b^*$ measurements were made after the first 28 hours (7 cycles) of total aging and then again after all 16 cycles were completed. Control specimens were stored under identical conditions but were not exposed to the artificial aging cycles.

Using the $L^*a^*b^*$ measurements total delta E (ΔE) was calculated comparing the values from the 28-hour and 64-hour accelerated artificial aging protocol to the baseline measurements. ΔE is a calculation of the total color change based on the three-dimensional axes L^* (light to dark) a^* (green to red) and b^* (blue to yellow). ΔE was calculated using the following equation:

$$\Delta E = [(L_2 - L_{baseline})^2 + (a_2 - a_{baseline})^2 + (b_2 - b_{baseline})^2]$$

A two-way ANOVA (factors: cement and measurement period) was performed with a post-hoc LSD test for pair-wise comparisons.

4. Results

The results for the control specimens are shown in Table 2. The color change measurements were all below a ΔE of 0.6 which is generally considered below the detection limit of the human eye. While R exhibited a trend for more color change there were no statistical differences in any of the cements after 7 or 16 cycles for the bulk or cement/ceramic assemblies not subjected to accelerated aging. The ΔE results for the bulk specimens are shown in Table 3. The two-way ANOVA showed that both cement and the number of aging cycles were significant factors in ΔE ($p=0.001$). The color change values for P, both shades of G and V were statistically similar to each other ($p>0.05$) and there was not a statistical difference for those cements between 7 cycles and 16 cycles. R generated a statistically higher change for ΔE compared to the other cements and that color shift was statistically greater for R after 16 cycles ($p<0.05$). The ΔE results for the cement/ceramic assemblies are shown in Table 4. The overall change in ΔE was less than what was observed with the bulk cement specimens. Even through the lithium disilicate ceramic wafer, R generated statistically higher change in ΔE than P, G and V ($p<0.05$).

The relative contribution of each constituent in its influence on the overall ΔE color change was dependent on the individual cement. In qualitative terms, P exhibited most change after aging in the b^* axis with a shift towards blue. G for both shades exhibited changes in L^* with a trend to lighter and b^* with a trend towards yellow. V exhibited a similar trend as G while R exhibited a trend to darker in the L^* axis and a strong trend to yellow in the b^* axis.

Discussion

There is an increased demand for esthetic dental restorative materials and that has led to numerous advances in dental ceramics for meeting patient's expectations. The color match to adjacent teeth as well as durability and color stability are critical elements in generating patient satisfaction [9]. With the development of higher strength more fracture resistant ceramics clinicians are afforded the chance to create more tooth conservative preparations for all-ceramic restorations. This is especially true for ceramic veneers placed on anterior teeth for esthetic reasons. With less ceramic bulk however there is more translucency in the final restoration making the contribution of the resin cement more critical for final shade and esthetics. If a cement is not color stable, esthetic failures may occur.

Delta E as calculated from the CIELAB $L^*A^*b^*$ measurements is a reflection of the overall change in the sensation of color; hue, value, and chroma. Using the CIELAB system has several advantages. For example, the 3D color space represented by the 3 axes includes are perceivable colors, enables color perception similar to the human eye and can quantify color parameters into numerical units. The model is also accurate, reproducible and data is able to be evaluated statistically.

There is however ongoing disagreements regarding the minimum perceivable color change that can be detected by the human eye. ΔE is the measure of change in visual perception of two given colors and is measured on a scale from 0 to 100 where 0 is no color difference and 100 is complete distortion. Schuessler [18] lists these standard perception ranges for human eye perception: ≤ 1.0 - not perceptible by the human eye, 1-2- perceptible by through close observation, 2-10- perceptible at a glance, > 10 - colors are demonstrably different. Some authors [19] note that while there are differences in sensitivity to color

Table 2: ΔE for Bulk and Cement/Ceramic Specimen Controls (no aging).

Cement	ΔE control-7 Bulk	ΔE -7 cycles Cement/Ceramic	ΔE control-16 Bulk	ΔE -16 cycles Cement/Ceramic
P	0.31 \pm 0.13	0.17 \pm 0.07	0.29 \pm 0.11	0.16 \pm 0.10
G A2	0.22 \pm 0.07	0.24 \pm 0.06	0.19 \pm 0.09	0.21 \pm 0.08
G Translucent	0.32 \pm 0.11	0.22 \pm 0.09	0.26 \pm 0.14	0.22 \pm 0.11
V	0.27 \pm 0.09	0.14 \pm 0.04	0.29 \pm 0.19	0.18 \pm 0.07
R	0.45 \pm 0.23	0.37 \pm 0.09	0.43 \pm 0.21	0.41 \pm 0.07

Table 3: ΔE for Bulk Cement Specimens after 7 and 16 aging cycles

Cement	ΔE AA-7 cycles	ΔE EAA 16 cycles
P	1.38 \pm 0.53 ^A	1.66 \pm 0.59 ^A
G A2	1.77 \pm 0.56 ^A	1.87 \pm 0.56 ^A
G Translucent	1.81 \pm 0.29 ^A	1.86 \pm 0.23 ^A
V	2.01 \pm 0.67 ^A	2.18 \pm 0.84 ^A
R	4.49 \pm 1.01 ^{B,*}	5.56 \pm 0.97 ^{B,*}

ΔE for the bulk cement specimens after 7 and 16 cycles of accelerated aging compared to baseline measurements. Cements connected by a similar capital letter were statistically similar within a vertical column. * denotes a significant difference for R between the 7 and 16 cycle ΔE aging intervals.

Table 4: ΔE for Cement/Ceramic Assemblies Specimens after 7 and 16 aging cycles.

Cement	ΔE AA-7 cycles	ΔE EAA 16 cycles
P	0.97 \pm 0.24 ^A	0.93 \pm 0.16 ^A
G A2	0.95 \pm 0.17 ^A	1.07 \pm 0.34 ^A
G Translucent	1.05 \pm 0.31 ^A	1.07 \pm 0.26 ^A
V	0.93 \pm 0.25 ^A	1.00 \pm 0.22 ^A
R	1.67 \pm 0.40 ^{B,*}	2.17 \pm 0.42 ^{B,*}

ΔE for the cement/ceramic specimens after 7 and 16 cycles of accelerated aging compared to baseline measurements. Cements connected by a similar capital letter were statistically similar within a vertical column. * denotes a significant difference for R between the 7 and 16 cycle ΔE aging intervals.

among individuals in the population that a ΔE difference between 2 and 3 units will be able to be perceived by most people. In the dental literature [21] some have proposed a ΔE of 3.3 to be the threshold for detection of color change.

The present study employed two kinds of accelerated aging factors. Water sorption due to the storage conditions can change the perception of color by changing the refractive index of the cement matrix [21]. The second accelerated aging factor was exposing the specimens to a xenon light source with a high intensity broad spectrum output. The absorption of light across the spectral range may result in degradation of both mechanical and chemical properties of the cement and catalyze physicochemical changes in the cement resulting in color changes in the material [22]. Specimens stored in water but not subjected to the xenon accelerated aging protocol exhibited minimal color changes and well below the threshold for human eye detection. Therefore, color changes observed in the bulk cements and as measured through the ceramic wafer were most likely affected by the accelerated aging process using the xenon light source.

Accelerated aging in the present investigation led to changes in ΔE for all experimental groups and increased the color parameter for RelyX Universal well past the critical threshold in the bulk specimens and likely near the detection limit for some human observers in the cement/ceramic group for RelyX. The color stability of RelyX was lower than that of all the other cements evaluated. The polymerization mechanisms of these dual-cured cements is complex and the degree of polymerization and the level of unreacted monomers is difficult to predict. In addition, the chemical composition of these cements is quite diverse, and the filler content and types of monomers used can have a significant effect on color stability.

Dual-cured cements are used in clinical situations where energy from the curing light may not be accessible [10]. Typically, dual-cured cements employ aromatic amines and benzoyl peroxide to drive the self-cure reaction while the light cured components include camphorquinone (CQ) as a photo-initiator and an aliphatic amine. The CQ/aliphatic amine systems are generally considered to be color stable while aromatic amines used with a peroxide catalyst tend to darken over time [7]. Recent investigations have confirmed that resin cements without benzoyl peroxide/amine initiators exhibit better color stability. Panavia V5 and G-CEM One are claimed by their respective manufacturers to have optimal color stability as a result of eliminating tertiary amines from their formulas. The result of this investigation would tend to support those claims. There are several limitations intrinsic to an in-vitro study of this kind. It is very challenging to simulate aging in the oral environment and the effects of other external factors such as plaque accumulation, alternations in pH and the effects of food discoloration were not represented here. In addition, just one thickness and one kind of dental ceramic was used in this investigation. Further studies will be required to more fully understand the effects of accelerated aging protocol on the color stability of resin cements used in the esthetic zone.

Conclusion

Within the limitations of this study the following conclusions can be made:

1. Water sorption was not a significant factor in color change of the cements for the time intervals used in this investigation.
2. Panavia V5, Variolink Esthetic and G-CEM One dual-cured cements showed acceptable color change in both bulk cement specimens and when evaluated with a 1mm lithium disilicate ceramic overlay.
3. RelyX Universal showed significantly higher color changes compared to the other cements in both bulk and when evaluated with the ceramic overlay.

Conflicts of Interest

The authors of this manuscript declare that they have no conflicts of interest, financial or nonfinancial in this article.

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References

1. Rosenstiel SF, Land MF, Crispin BJ. (1988). Dental luting agents: a review of the current literature. *Journal of Prosthetic Dentistry*. 80: 280-301.
2. Heffernan MJ, Aquilino SA, Diaz-Arnold AM, Haselton DR, Stanford CM, et al. (2022). Relative translucency of six all-ceramic systems. Part II. Core and veneer materials. *Journal of Prosthetic Dentistry*. 88: 10-5.
3. Nathanson D, Banasr F. (2002). Color stability of resin cements - an in vitro study. *Practical Procedures and Aesthetic Dentistry*. 14: 449-455.
4. Valizadeh S, Asiaie Z, Kiomarsi N, Kharazifard MJ. (2020). Color stability of self-adhering composite resins in different solutions. *Dent Med Probl*. 57: 31-38.
5. Perroni AP, Bergoli CD, Dos Santos MBF, Moraes RR, Boscato N. (2017). Spectrophotometric analysis of clinical factors related to the color of ceramic restorations: A pilot study. *J Prosthet Dent*. 118: 611-616.
6. Saati K, Valizadeh S, Anaraki SN, Moosavi N. (2021). Effect of aging on color stability of amine-free resin cement through the ceramic laminate veneer. *Dent Res J*. 18: 99.
7. Atay A, Palazli Z, Gürdal I, Üşümez A. (2019). Color change of different dual-cure resin cements after thermocycling. *Odovtos Int J Dent Sci*. 21: 53-62.
8. Janda R, Roulet JF, Latta M, Steffin G, Ruttermann S. (2005). Color stability of resin-based filling materials after aging when cured with plasma or halogen light. *European Journal of Oral Sciences*. 113: 251-257.
9. Alkurt M, Duymus ZY. (2018). Comparison to color stability between amine with benzoyl peroxide includes resin cement and amine-reduced, amine-free, lacking of benzoyl peroxide resin cements after thermocycle. *J Adv Oral Res*. 9: 24-30.
10. Catelan A, Suzuki TY, Becker F Jr., Briso AL, Dos Santos PH. (2017). Influence of surface sealing on color stability and roughness of composite submitted to ultraviolet-accelerated aging. *J Investig Clin Dent*. 8: 1-5.
11. Ertas E, Guler AU, Yucel AC, Koprulu H, Guler E. (2006). Color stability of resin composites after immersion in different drinks. *Dental Materials Journal*. 25: 371-376.
12. Ramos N, Luz J, Valera M, Melo R, Saavedra G, et al. (2019). Color Stability of Resin Cements Exposed to Aging. *Operative Dentistry*. 44: 609-614.
13. Kilinc E, Antonson S, Hardigan P, Kesercioglu A. (2011). Resin cement color stability and its influence on the final shade of all-ceramics. *Journal of Dentistry*. 39s: e30-e36.
14. Ataie M, Fayaz A, Ghasemi A, Sheikh-Al-Eslamian SM, Farahani S. (2023). Color stability of three commercial resin cements after accelerated aging. *J Conserv Dent*. 26: 212-215.
15. Lei MA, Rivelli M, Iglesias AM, Marquez JG, Gonzalez N, et al. (2022). Accelerated artificial aging and color stability in resin-based cements. *Acta Odontol Latinoam*. 35: 67-73.
16. Mazzitelli C, Paolone G, Sabbagh J, Scotti N, Vichi A. (2023). Color Stability of Resin Cements after Water Aging. *Polymers (Basel)*. 15: 655.
17. Ozkanoglu S, G Akin EG. (2020). Evaluation of the effect of various beverages on the color stability and microhardness of restorative materials. *Niger J Clin Pract*. 23: 322-328.
18. (2024). Zachary Schuessler, *Delta E* 101.

19. Brainard Dh. (2003). Color Appearance and Color Difference Specification. In *The Science of Color*. 192- 213.
20. Bayindir F, Koseoglu M. (2020). The effect of restoration thickness and resin cement shade on the color and translucency of a high-translucency monolithic zirconia. *J Prosthet Dent*. 123: 149-154.
21. Sideridou I, Achilias DS, Spyroudi C, Karabela M. (2004). Water sorption characteristics of light-cured dental resins and composites based on Bis-EMA/PCDMA. *Biomaterials*. 25: 367-376.
22. Turgut S, Bagis B. (2011). Colour stability of laminate veneers: an *in vitro* study. *J Dent*. 39: e57-64.