The effect of staining solutions on the color stability of the provisional restorative materials

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Objective: To investigate color stability of four provisional restorative materials after immersion in different staining solutions at various immersion time.

Materials and methods: Four provisional restorative materials were selected, namely, methacrylate resin (Unifast Trad) and bis-acryl resins (Protemp 4, Luxatemp Fluorescence and Integrity). Twenty-one disc shaped specimens (15 mm diameter, 1 mm thick) were prepared from each provisional restorative materials according to the manufacturer's instruction. Seven specimens of each material were randomly assigned for immersion in distilled water (control), coffee and curry. All specimens were stored in the solutions at 37°C. According to CIELAB system, the color baseline was measured with the spectrophotometer. The color change (ΔE) was measured after immersion for 7, 30 and 90 days. Data were analyzed statistically with two-way repeated analysis of variance and multiple comparisons. Results: Distilled water (control): All provisional restorative materials had no statistically significant difference in mean color change at day 7 (p>0.05). At day 30, color change of Unifast Trad (ΔE_{30} =1.5±0.18) was significantly higher than Luxatemp ($\Delta E_{30} = 0.80 \pm 0.52$) (p<0.05). At day 90, Unifast Trad ($\Delta E_{30} = 3.58 \pm 0.29$) showed significantly higher color change than others (p<0.05). Coffee: All provisional restorative materials had no statistically significant difference in color change at day 7 and 90 (p>0.05), except Integrity ($\Delta E_7 = 11.93 \pm 4.69$), $(\Delta E_{90} = 21.80 \pm 3.88)$ which had significantly higher color change (p<0.05). At day 30, Integrity ($\Delta E_{30} = 15.81 \pm 4.17$) showed significantly higher color change than Luxatemp (ΔE_{so} = 11.53±2.91) and Unifast Trad (ΔE_{so} = 10.39±1.10) (p<0.05). Curry: All provisional restorative materials had no statistically significant difference in color change at day 7 and 30, except Unifast Trad (ΔE_7 =6.14±0.98), (ΔE_{30} =9.32±3.17) which had significantly lower color change (p<0.05). At day 90, Unifast Trad (ΔE_{00} =24.41±3.37) showed significantly lower color change than others (p<0.05). Conclusion: The type of provisional restorative materials, staining solutions and immersion time could affect the color stability of provisional restorative materials.

Key words: Bis-acryl resin, color change, color stability, methacrylate resin, provisional restorative materials, staining solutions

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Introduction

Provisional restorations play an important role in fixed partial prosthodontics. They should provide pulpal protection, positional stability, maintenance of occlusal function, cleanability, high strength, reliable retention and esthetics. [1] Provisional restorations should not only provide

an initial shade match but also maintain the shade over the provisionalization period until the definitive prostheses have been fabricated. In some cases, such as evaluation of periodontal treatment, alveoloplasty, oral rehabilitation, dental implant placement, endodontic treatment and orthodontic treatment, long-term provisionalization is indicated.

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Methacrylate resins have been used as provisional restorations for years because of their high strength, low cost, easy handling and repair. However, the disadvantages of the resins are unpleasant odor, high temperature exothermic setting reaction, and direct cytotoxic effects of monomers to dental pulp. In recent years, bis-acryl resins have been introduced to the market. It is based on multi-functional methacrylic esters similar to the resin composites. Bis-acryl resin materials offer the advantages of low temperature exothermic reaction, low pulpal irritation and better marginal adaptation but it is difficult to repair and more expensive than the methacrylate resin. [2, 3]

Color stability of provisional restoration is a significant physical property. It describes the ability of materials that can maintain their initial shade. This property is influenced by patient's habits as well as surface smoothness, chemical and physical properties of materials. [4] The degree of color change may depend on incomplete polymerization, water sorption, surface smoothness, chemical reaction, material composition, types of immersion solution and the amount of exposure time. [5]

Many in vitro experimental studies [4, 6-9] were conducted to evaluate the discoloration of resin composites, acrylic resins and denture bases immersed in the staining solutions, but the number of studies about the discoloration of bis-acryl resins is limited. In most previous studies [5, 10, 11], the provisional restorative materials were exposed to staining solutions for a short period of time.

The objective of this study was to investigate color stability of provisional restorative materials after immersion in different staining solutions for long period of immersion time.

Materials and methods

In this study, four provisional restorative materials (Unifast Trad, Protemp 4, Luxatemp Fluorescence and Integrity) were selected and shown in Table 1. The total of twenty-one disc shaped specimens (15 mm diameter, 1 mm thick) were fabricated from each provisional restorative material using a metallic mold. A piece of polyvinyl film was placed on a glass slab and the metallic mold applied with a mold-release agent was placed on it. Then, provisional restorative materials were mixed according to the manufacturer's recommendations and cautiously filled into the metallic mold. Another piece of polyvinyl film was placed on the material and covered with a glass slab on top of the mold. The finger loading was pressed against it until the materials set. After removing the specimens from the mold, they were wet finished by using a sequence of silicon carbide abrasive papers grit no.800 and 1,000 (TOA, Samut Prakan, Thailand) and speed of 150 rpm in a polishing machine (Struers Model RotoForce-4, USA). The dimension of the specimens was measured by the digital caliper of the accuracy 0.01 mm. All specimens were stored in distilled water at 37°C for 24 hours. The color baseline of all specimens were measured using the spectrophotometer (Ultrascan Colorquest XE, HunterLab, USA). The color of the specimens were assessed in the Commission International de' l'Èclairage (CIE) L*a*b* color system. [12] L* is lightness; white to black, a* is red-green axis ; positive value indicates red, negative value indicates green and b* is yellow-blue axis; positive value indicates yellow, negative value indicates blue.

The specimens were randomly separated into distilled water (control), coffee and curry. Staining solutions were prepared according to manufacturer's instruction (Table 2). All specimens were immersed in solutions at 37°C over a 90 days test period. The solutions were renewed every three days. A color measurement of CIE L*a*b* using the spectrophotometer was performed by one investigator after 7, 30 and 90 days of immersion. The total color change (ΔE) of each test specimen was calculated using this equation:

$$\Delta E = (\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2})^{1/2}$$

where ΔL^* , Δa^* , Δb^* are the differences between color baseline L*a*b* and L*a*b* after immersed in solutions for 7, 30 and 90 days. Prior to the color measurement, the spectrophotometer was calibrated according to the manufacturer's recommendation.

Provisional restorative materials used in this study

Materials	Types	Manufacturers	Compositions	Filler size	Shade
GC Unifast Trad [™]	Metha crylate	GC America, Illinois, USA	Powder: Methyl methacrylate and Ethyl methacrylate copolymer Liquid: Methyl methacrylate, butylated hydroxytoluene, hydroquinone	-	lvory
Protemp [™] 4	Bis-acryl	3M ESPE, Seefeld, Germany	Base paste: Dimethacrylate(BisEMA6), Silane treated amorphous silica, Reaction production products of 1,6-diisocyanatohexane with 2-{(2-methacryloyl)ethyl]6-hydroxyhexanoate and 2-hydroxyethylmethacrylate (DESMA), Silane treated silica Catalyst paste: Ethanol, 2,2'- [(1-methylethylidene)bis(4,1-phenyleneoxy)] bis-, diacetate, Benzyl-phenyl-barbituric acid, Silane treated silica, Tert-butyl peroxy- 3,5,5-trimethylhexanoate	20-30 nm	A3
Luxatemp Fluorescence [®]	Bis-acryl	DMG, Hamburg, Germany	Base paste: Acrylic resin glass powder silica Catalyst paste: Urethane dimethacrylate, Aromatic dimethacrylate, Glycol methacrylate	3 μm	А3
Integrity TM	Bis-acryl	Dentsply Caulk, Delaware, USA	Barium boron alumino sillicate glass, Hydrophobic amorphous fumed silica, methacrylate monomers, Polymerizable dimethacrylate resin, Catalyst, Stabilizers	3 μm	A3

Table 2 Staining solutions

Staining solutions	Manufactures	Preparations
Coffee	Nescafe red cup, Nestle, Thailand	4 g of instant coffee and 240 ml of hot water
Curry	Yellow curry paste, Kanokwan Food Products	50 g of yellow curry paste and 480 ml of
	Co., Ltd, Thailand	hot water

Results

The distribution of color change data analyzed using Shapiro-Wilk test was normal. Levene's test showed the homogeneity among the group variances. Then, the color change data were analyzed using two-way analysis of variance by repeated measurement and multiple comparisons: Bonferroni in statistical package for the social sciences 18.0 (SPSS 18.0, SPSS Inc. Chicago, Illinois, USA).

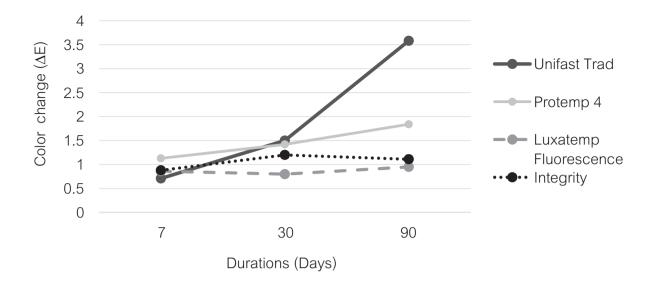
In distilled water, the mean color change and standard deviations are shown in Table 3 and Figure 1. There was no statistically significant difference in mean color change (ΔE) of all provisional restorative materials at day 7 (p>0.05). At day 30, color change of Unifast Trad (ΔE_{30} = 1.50±0.18) was significantly higher than Luxatemp $(\Delta E_{30} = 0.80 \pm 0.52)$ (p<0.05) while no significant difference was found among Unifast Trad $(\Delta E_{30} = 1.50 \pm 0.18)$, Protemp 4 $(\Delta E_{30} = 1.42 \pm 0.55)$ and Integrity ($\Delta E_{30} = 1.20 \pm 0.37$) (p>0.05). At day 90, Unifast Trad ($\Delta E_{90} = 3.58 \pm 0.29$) showed significantly higher color change than others (p<0.05). Luxatemp (ΔE_{so} = 0.95±0.41) and Integrity (ΔE_{90} =1.11±0.30) had no significant difference in color change (p>0.05) but both materials had significantly lower color change than Protemp 4 ($\Delta E_{90} = 1.84 \pm 0.61$) (p<0.05). In addition, Unifast Trad exhibited significantly greater color change at all periods of immersion time (p<0.05) while Protemp 4 exhibited significantly greater color change only at day 90 (p<0.05). On the contrary, Luxatemp and Integrity showed no significant color change at all periods of immersion time (p>0.05).

In coffee, the mean color change and standard deviations are shown in Table 4 and Figure 2. There was no statistically significant difference in color change of all provisional restorative materials at day 7 and 90 (p>0.05), except Integrity which had higher color change $(\Delta E_7 = 11.93 \pm 4.69)$, $(\Delta E_{90} = 21.80 \pm 3.88)$ than others at day 7 and 90 (p<0.05). Moreover, at day 7 and 90 there was no significant color change among Protemp 4 ($\Delta E_7 = 5.26 \pm 1.33$), ($\Delta E_{90} = 16.69 \pm 3.18$) Luxatemp ($\Delta E_7 = 5.86 \pm 2.15$), ($\Delta E_{qq} = 16.40 \pm 2.77$) and Unifast Trad ($\Delta E_7 = 3.89 \pm 1.24$), ($\Delta E_{00} =$ 16.42±2.38) (p>0.05). At day 30, no significant difference in color change was found among Unifast Trad (ΔE_{30} =10.39±1.10), Protemp 4 (ΔE_{30} = 11.89 \pm 2.16) and Luxatemp (ΔE_{30} =11.53 \pm 2.91) (p>0.05). However, Integrity ($\Delta E_{30} = 15.81 \pm 4.17$) showed significantly higher color change than Luxatemp (ΔE_{30} =11.53±2.91) and Unifast Trad $(\Delta E_{30}=10.39\pm1.10)$ (p<0.05) but not different from Protemp 4 (ΔE_{30} =11.89±2.16) (p>0.05). Further, all provisional restorative materials exhibited significantly greater degree of color change at all periods of immersion time (p<0.05).

Mean color change (ΔE), standard deviations (SD) of provisional restorative materials after immersion in distilled water for different periods of time

	Durations (days)						
Materials	7		30	30		90	
	ΔΕ	SD	ΔΕ	SD	ΔΕ	SD	
Unifast Trad	0.71 ^{a, A}	0.30	1.50 a, B	0.18	3.58 a, C	0.29	
Protemp 4	1.13 ^{a, A}	0.79	1.42 ab, A	0.55	1.84 b, B	0.61	
Luxatemp Fluorescence	0.86 a, A	0.69	0.80 b, A	0.52	0.95 c, A	0.41	
Integrity	0.88 a, A	0.36	1.2 ab, A	0.37	1.11 c, A	0.30	

^{*}The different capital letters in the same row and lower-case letters in the same column represent dissimilar color change of provisional restorative materials at 5% level of significance (p<0.05) by Bonferroni multiple comparisons.

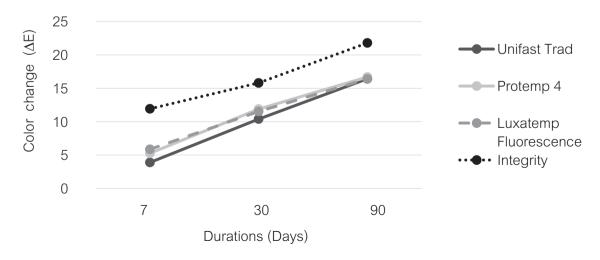


Mean color change (ΔE) of provisional restorative materials after immersion in distilled water

Table 4 Mean color change (ΔE), standard deviations (SD) of provisional restorative materials after immersion in coffee for different periods of time

	Durations (days)						
Materials	7		30	30		90	
	ΔΕ	SD	Δ E	SD	Δ E	SD	
Unifast Trad	3.89 a, A	1.24	10.39 a, B	1.10	16.42 a, C	2.38	
Protemp 4	5.26 a, A	1.33	11.89 ab, B	2.16	16.69 a, C	3.18	
Luxatemp Fluorescence	5.86 a, A	2.15	11.53 a, B	2.91	16.40 a, C	2.77	
Integrity	11.93 b, A	4.69	15.81 b, B	4.17	21.80 b, C	3.88	

^{*}The different capital letters in the same row and lower-case letters in the same column represent dissimilar color change of provisional restorative materials at 5% level of significance (p<0.05) by Bonferroni multiple comparisons.



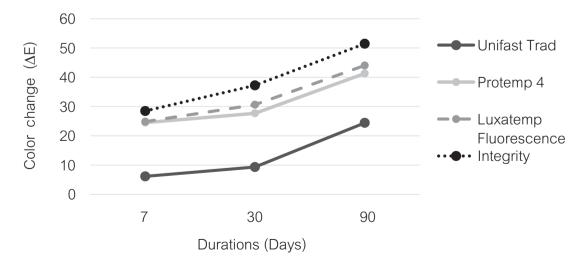
Mean color change (ΔE) of provisional restorative materials after immersion in coffee.

In curry, the mean color change and standard deviations are shown in Table 5 and Figure 3. Unifast Trad ($\Delta E_7 = 6.14 \pm 0.98$), ($\Delta E_{30} =$ 9.32±3.17) showed significantly lower color change (p<0.05) at day 7 and 30 while no significant difference was found among Protemp 4 (ΔE_7 = 24.50 \pm 10.28), (ΔE_{30} =27.68 \pm 20.28), Luxatemp $(\Delta E_7 = 24.83 \pm 4.46)$, $(\Delta E_{30} = 30.62 \pm 9.02)$ and Integrity ($\Delta E_7 = 28.47 \pm 4.31$), ($\Delta E_{30} = 37.22 \pm 7.83$) (p>0.05). At day 90, Unifast Trad (ΔE_{qq} =24.41±3.37) showed significantly lower color change (p<0.05). Integrity (ΔE_{oo} =51.45±5.08) showed significantly higher color change than Unifast Trad (ΔE_{on} = 24.41 \pm 3.37) and Protemp 4 ($\Delta E_{00} = 41.29 \pm 9.62$) (p<0.05). No significant difference in color change was found between Protemp 4 (ΔE_{90} =41.29±9.62) and Luxatemp ($\Delta E_{qq} = 44.02 \pm 5.33$) (p>0.05) and between Luxatemp ($\Delta E_{on} = 44.02 \pm 5.33$) and Integrity ($\Delta E_{90} = 51.45 \pm 5.08$) (p>0.05). In addition, Unifast Trad and Protemp 4 showed significantly greater color change at day 90 (p<0.05) while Luxatemp and Integrity exhibited significantly greater degree of color change at all periods of immersion time (p<0.05).

Table 5 Mean color change (ΔE), standard deviations (SD) of provisional restorative materials after immersion in curry for different periods of time

	Durations (days)						
Materials	7		30		90		
	ΔΕ	SD	ΔΕ	SD	ΔΕ	SD	
Unifast Trad	6.14 a, A	0.98	9.32 a, A	3.17	24.41 a, B	3.37	
Protemp 4	24.50 b, A	10.28	27.68 b, A	10.28	41.29 b, B	9.62	
LuxatempFluorescence	24.83 b, A	4.46	30.62 b, B	9.02	44.02 bc, C	5.33	
Integrity	28.47 b, A	4.31	37.22 b, B	7.83	51.45 ^{c, C}	5.08	

^{*}The different capital letters in the same row and lower-case letters in the same column represent dissimilar color change of provisional restorative materials at 5% level of significance (p<0.05) by Bonferroni multiple comparisons.



Mean color change (ΔE) of provisional restorative materials after immersion in curry.

Discussion

Color stability is a significant physical property of provisional restorations. It is defined as the ability of materials that can maintain their initial shade. In this experiment, the color change (ΔE) of methacrylate resins (Unifast Trad) and three bis-acryl resins (Protemp 4, Luaxatemp Fluoresence and Integrity) were investigated after immersion in various staining solutions for long duration (7, 30 and 90 days).

Color measurement

Color change assessed by visual perception is subjective. Physiologic and psychologic factors are involved in the process. In this study, the Commission International de'l'Èclairage (CIE) LAB color system [12] was used to assess color change to eliminate the subjective error of the visual color assessment. [13]

The ΔE represents color change between the color at baseline and after immersion in solutions. Discoloration of provisional restorative materials is a problem for patients and clinicians. Darkening of provisional restorations when using in the longterm treatments as full mouth rehabilitation, periodontal treatment and orthodontic treatment can lead to unsatisfied esthetics especially in anterior region during period of treatment. Johnson and Kao [13] evaluated color matching in composite resin veneers of patient teeth by using two visual rating systems such as United Stated Public Health Service (USPHS) and extended visual rating scale for appearance match (EVRSM) and instrument colorimetry using CIELAB system. They reported that the clinically acceptable ΔE was about 3.7. In this study, all provisional restorative materials immersed in distilled water for long period showed color change (ΔE) below 3.7 while ΔE after being immersed in coffee and curry at the same period were above 3.7. Therefore, provisional restorative materials exhibited clinically

unacceptable color change when immersion in coffee and curry at long period of time.

Staining solutions

The different colorant agents selected for staining provisional restorative materials in this experiment were coffee and curry. The absorption and adsorption of different polarities of colorant agent influence color change of provisional restorative materials. Coffee has a brown pigment known as tannin which has ability to stain teeth and provisional restoratives. Tannin are compounds of intermediate to high molecular weight and can form insoluble complex with carbohydrates and protein. [14] Curry is composed of several spices and oil substance. Its yellowish appearance is the most common recognition. Turmeric which is the main spice of curry, contains yellow chemical product known as curcumin. Curcumins contain poly-phenols compounds which are non-polar and cannot dissolve in water. [14] Borges et al. [15] demonstrated that the degree of polarity of dyes determines their degree of penetration into the resins. Low polar dyes, such as coffee can penetrate easily into the polymer matrix while more polar dyes such as wine only impregnate the surface of material. Therefore, non-polar dyes such as curcumin penetrate more easily into the polymer matrix. Moreover, pH can affect the degree of dye penetration of provisional restorative materials. Erdemir at al. [16] reported that a low pH can negatively affect the surface integrity by softening the matrix due to loss of structure ions such as calcium, aluminium, silicon and increasing the pigment absorption. Initial pH of staining solutions was measured in this experiment using pH meter (ORION 3-star, Expotech, USA). The pH of tested curry, coffee and distilled water were 4.74, 5.25 and 7, respectively. All provisional restorative materials immersed in coffee exhibited lower color change than in curry and higher color change than in distilled water. Therefore, the color change of all

provisional restorative materials ranged between 6.14 - 51.45 in curry while in coffee, color change ranged between 3.89 -21.80 and in distilled water color change ranged between 0.71- 3.58. However, not only pH and polarity of food colorant that were factors associated color change of provisional restorative materials but different compositions of provisional restorative materials were also significant factors. In present study, specimens stained by coffee showed greater a* (red-green) value while those stained by curry showed greater b* (yellow-blue) value.

Provisional restorative materials

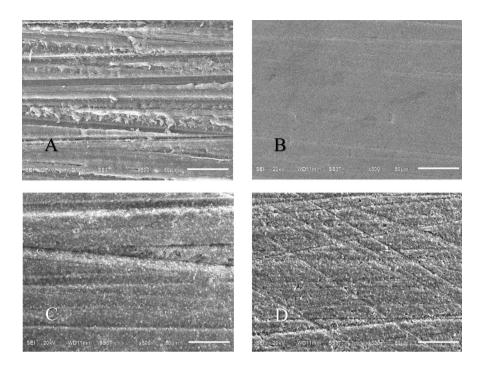
In distilled water, at day 90 Unifast Trad (methacrylate resins) exhibited significantly higher color change (ΔE) than three bis-acryl resins (Protemp 4, Luxatemp and Integrity). At day 30, Unifast Trad exhibited significantly higher color change than Luxatemp. The result of this investigation was similar to the study of Sham et al¹¹ who studied five provisional restorative materials by immersing the specimens into water at 37°C for 20 days. Methacrylate resins showed significantly higher color change than the bis-acryl resins.

In coffee, at day 7, 30 and 90 color change of Unifast Trad (methacrylate resins) and two bis-acryl resins were not significantly different except Integrity which showed higher color change than other provisional restorative materials. Bayindir et al. [5] also found that bis-acryl resins showed significantly higher color change than methacrylate resins after immersed in coffee at 37°C for 7 and 30 days. Furthermore, Sham et al. [11] discovered that bis-acryl resins showed significantly higher color change than methacrylate resins after immersed in coffee at 37°C for 20 days. In present study, higher color change of Integrity may be influenced by their larger filler particles (Table1) and higher surface roughness than others. Composite resins with large filler particles are more prone to water aging discoloration than

composite resins with small filler particles because they are susceptible to the hydrolytic degradation of matrix and filler interfaces. [16] Moreover, large filler particles produce high surface roughness. [18]

In this study, additional observation of the surface texture was also performed to investigate the surface smoothness and porosity of the tested materials. All specimens were prepared using the same procedures as the tested specimens. Then, the specimens were cleaned in the ultrasonic machine. The samples were examined by scanning electron microscopy (JSM 6610LV, JEOL, JAPAN) (Figure 4). The scanning electron microscopic examination showed various surface characteristics of provisional restorative materials: Unifast Trad presented numerous deep and large scratches, Protemp 4 presented smooth surface with few shallow scratches, Laxatemp Fluorescence presented numerous deep, large scratches and porous structure, Integrity presented numerous deep narrow scratches and porous structure. Remarkably, Integrity had more flaws and scratches than other bis-acryl resins leading to higher color change when immersed in extrinsic staining solutions such as coffee. Crispin and Caputo [19] showed that materials with rough surface could generate darker color more than smooth surface materials. In addition, Haselton et al. [20] showed baseline surface roughness measurement of provisional crown and fix partial denture. Integrity had higher surface roughness than Luxatemp, Protemp Garant and Unifast LC.

In curry, at day 7, 30 and 90 Unifast Trad showed significantly lower color change than three bis-acryl resins. At day 7 and 30 no significant difference was found among three bis-acryl resins. This result was similar to the result of Watanabe et al. [4] The color change was evaluated between methacrylate resin and two bis-acryl resins after immersion in curry for 14 days. Methacrylate resins exhibited less color change than bis-acryl resins after 14 days.



SEM image of surface of each provisional restorative materials before immersion in solutions (x500), Figure 4 scale bar 50 µm: (A) Unifast Trad presented numerous deep and large scratches; (B) Protemp 4 presented smooth surface with few shallow scratches; (C) Luxatemp Fluorescence presented numerous deep, large scratches and porous structure; (D) Integrity presented numerous deep narrow scratches and porous structure.

In this study, all provisional restorative materials were immersed in different staining solutions such as coffee and curry for long period. Bis-acryl resins showed higher color change than methacrylate resins. This may be due to their higher diffusion coefficient when compared to methacrylate resins and no filler contained in methacrylate resins. Therefore, bis-acryl resins exhibited higher water sorption leading to the higher color change. [21] Another factor that influences higher color change of bis-acryl resins is their compositions. They contain bis-EMA (ethoxylated bisphenol-A dimethacrylate), UDMA (urethane dimethacrylate), and bis-GMA (bisphenylglycidyl dimethacrylate) as the resin matrix. Bis-GMA monomer tends to be hydrophilic, contains polar hydroxyl group and can form hydrogen bond with water. Previous studies [17, 22] showed that bis-EMA has less susceptible to water sorption than bis-GMA and UDMA is less water sorption than bis-GMA. If the material has

ability to absorb water, it can also absorb the colorant agent. Protemp 4 presents bis-EMA as resin matrix and it presents lower color change when immersion in curry at all immersion periods.

Immersion time

At 90 days of immersion time in all staining solutions, all provisional restorative materials showed significantly greater color change when compared to baseline color except Luxatemp and Integrity which showed no significant color change at all periods of immersion in distilled water. The color change was affected by water sorption characteristics. Long immersion time may cause excessive water sorption and decrease lifetime of composite resins by expanding and plasticizing the resins component, hydrolyzing the silane and causing micro-crack formation or interfacial gaps at the interface between the filler and matrix. This could allow stain penetration or discoloration of provisional restorative materials. [16]

The color change of provisional restorative materials depends on compatibility of colorant agents and immersed materials. [8] Further studies regarding color stability of provisional restorative materials after thermocycling should be investigated. Also, the improvement of surface texture of provisional restorative materials should be considered.

Conclusion

Under the conditions for this study, it can be concluded that the type of provisional restorative materials, the staining solutions and immersion times could affect color stability of provisional restorative materials. When all provisional restorative materials are exposed to staining solutions, color change can be noticeably visible especially long-term exposure. Food colorants such as curry and coffee should be avoided.

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