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Research Article

Colour Stability of Restorative Materials: A Spectrophotometric Study

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A R T I C L E   I N F O

A B S T R A C T

Background: Colour stability of esthetic restorative materials is a crucial factor for restorations success. The aim of this study to determine the colour stability of two newly introduced composite resins after artificial aging in coffee and black tea.

Materials and Methods: Disk-shaped specimens from each resin composite (Ceram X one, Dentsply, Germany and G-aenial, GC Corporation, Japan) were prepared and immersed in coffee, tea or distilled water for 24hours and one month. Colour measurements were made at baseline, 24hours and one-month intervals using a reflectance spectrophotometer (UV-VIS 2401PC, SHIMADZU, Japan) and the CIEDE2000 system.

Statistical Analysis: Mean values and standard deviation were figured out for each specimen and data were statistically analysed with the use of IBM® SPSS® Statistics for Windows, Version 25.

Results: For both time periods of immersion in coffee, there were no statistically significant differences between mean ΔE values of the tested materials (P>0.05). One month after immersion in tea, ΔE values of Ceram X one and G-aenial presented statistically significant difference (P<0.05).

Conclusion: Coffee showed the greatest staining potential, followed by black tea. After one-month immersion in coffee, ΔE was material independent. Conversely, after one-month aging in tea, the colour difference ΔE was material and time dependent.

Introduction

Resin-based composites have evolved greatly in the last decades and they are one of the materials of choice for anterior restorations, so these materials need to demonstrate not only physicomechanical but also esthetic properties [1-4]. Esthetics of these materials is regulated by opacity, translucency, opalescence, fluorescence, colour stability, surface gloss and roughness [1, 5, 6]. Colour alteration of the resin-based materials occurs due to intrinsic discolouration or extrinsic staining [5-9]. Oral environment could be negatively influenced by bacterial activity or acidic drinks and foods. Since acid environment can alter roughness, hardness, flexural properties and fluorescence intensity of composite dental materials [10-13]. Water sorption, or adsorption of food

colourants such as red wine, coffee, coke, tea, UV irradiation and degree of polymerization could cause extrinsic staining of resin materials [1, 7-9, 14-19]. While intrinsic discolouration is defined as the staining of the resin material itself and is associated with the type of the resin matrix and the fillers' size and distribution [1, 6, 8, 9, 14, 16].

Studies have reported that materials with urethane dimethacrylate matrix are more stain resistant compared to materials with bis-GMA matrix because UDMA demonstrates lower viscosity and water absorption [9, 16]. Siloranes have been suggested as alternatives to methacrylates as matrix resin components for resin-based composites due to their hydrophobicity, decreased water sorption, solubility and lower polymerization shrinkage [20-24]. These materials seem promising in

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order to overcome discolouration from fluids in the oral environment. Several colour measuring systems have been proposed by the CIE, the most commonly used colour measuring system is CIE L*a*b* or CIE76 system. However, the most recent and officially recommended as the new colour difference equation is the CIEDE2000 system. This formula is designed in order to improve the performance for blue and gray colours and includes weighting functions of colour parameters [25].

Since new materials have been introduced for restorations in the anterior zone, which requires high esthetics, the purpose of this study was to determine the colour stability of two contemporary composite resins, recommended for anterior restorations, after artificial aging in coffee and tea solutions by using a spectrophotometer device.

Materials and Methods

Three different solutions (coffee, tea and distilled water) were used to evaluate their effect on colour stability of two composite materials (Ceram X one, Dentsply, Konstanz, Germany and G-aenial, GC Corporation, Tokyo, Japan). The composition of tested materials is shown in (Table 1).

Table 1: Characteristics & contents of composite resins used in this study.

Product	Shade	Composite Resin Type	Synthesis	Manufacturer
Ceram X One Universal	A2	Nanohybrid	Polysiloxane, Poly-urethane-methacrylate, bis-EMA, TEGDMA	Dentsply, Detrey GmbH, Konstanz, Germany
G-aenial Anterior	A2	Hybrid	UDMA, dimethacrylate, co-monomers	GC Corporation, Tokyo, Japan

I Preparation of Staining Solutions

Coffee (Jacobs, Douwe Egberts, 1011 DK Amsterdam, The Netherlands) was prepared automatically at a coffee maker with the proportion of three spoons of coffee per one cup of water (300 ml) according to manufacturer's instructions. For the black tea (Yellow Label® Black Tea, Lipton®, Unilever) solution preparation, there were used one tea bag per one cup of boiling water (300ml), while the time was settled on three minutes, according to manufacturer's instructions.

II Specimens Preparation

Thirty (30) specimens of each resin composite were prepared using standardized Teflon molds (diameter: 10mm and height: 2mm) according to the manufacturer's instructions. Number of specimens was calculated according to power analysis. The mold with the composite resin was held between two glass slides, the slides were then gently pressed together to remove excess material [4]. All specimens were polymerized (Acteon® Satelec, France) with light intensity of 1200 mW/cm^2 according to manufacturer's instruction with the light tip 1mm away from the specimen [4, 26]. After polymerization the specimens were stored at an incubator with constant conditions (37ºC and 100% humidity) for 24 hours so that polymerization process could be completed. Afterwards, polishing was conducted via silicon carbide finishing papers of decreasing grit sizes: P180, P220, P240 for both sides of specimens. Following that, the samples were rinsed with running water for one minute and were blotted dry with absorbent paper. The specimens were randomly divided into three groups (n=10), immersed in the solutions (coffee, tea and distilled water) and stored at an incubator with constant conditions (37ºC and 100% humidity) for 24 hours and one month. Staining solutions were renewed every two days to avoid bacteria or yeast contamination [4, 27].

III Determination of Colour Stability

Measurements were performed with the use of a reflectance spectrophotometer (UV-VIS 2401PC, SHIMADZU, Japan) with CIEDE 2000 system. Before measurements the spectrophotometer was calibrated according to manufacturer 's guidelines by using the supplied white calibration standard (Barium sulfate: BaSO$_4$ base) and the wavelength range was set among: 780nm to 380nm. Before specimens' immersion baseline colour measurements were made. Colour stability was measured using the CIEDE 2000 colour difference (ΔE_{00}) according to the following formula:

$$\Delta E_{00} = \sqrt{\left(\frac{\Delta L'}{K_L S_L}\right)^2 + \left(\frac{\Delta C'}{K_C S_C}\right)^2 + R_T \left(\frac{\Delta C'}{K_C S_C}\right)\left(\frac{\Delta H'}{K_H S_H}\right)}$$

where $\Delta L'$, $\Delta C'$, and $\Delta H'$ are the mathematical differences in lightness (L), chroma (C), and hue (H), respectively, between the two measurement periods and R_T is the rotation factor that accounted for interactions between chroma and hue differences in the blue region. Weighting functions, S_L, S_C, and S_H adjust the total colour difference for variation in the location of the colour difference pair in L, a, and b coordinates, and the parametric factors, K_L, K_C, and K_H, are correction terms for experimental conditions. Colour measurements were then made according to the same procedure at a time interval of 24 hours and one month.

IV Statistical Analysis

Mean values and standard deviation were figured out for each specimen and data were statistically analysed with the use of a software (IBM® SPSS® Statistics for Windows, Version 25, 64-bit Edition). Initially a test of normality was conducted to check whether the results follow normal distribution or not. Afterwards, mean values (ΔE) were compared using independent samples T-test, Levene's test of homogeneity of variance, paired sample T-test. The significance level was set at: P=0.05.

Results

The means values and standard deviation of colour change and colour parameters are displayed in (Table 2). Coffee seemed to have the greatest staining potential among the used solutions, since both materials presented statistically significant higher ΔE_{00} values for both observation times. No statistically significance differences between mean ΔE_{00} values of Ceram X one Universal and G-aenial anterior for both time periods, when the specimens were immersed in coffee solution (P>0.05). 24 hours after immersion in tea, there was also no statistically significant

difference between mean ΔE_{00} values of Ceram X one Universal and G-aenial anterior (P>0.05). While one month after immersion in tea, ΔE_{00}

values of Ceram X one Universal were statistically significantly lower than ΔE_{00} values of G-aenial anterior (P<0.05).

Table 2: Means and standard deviations of colour change (ΔEab) ΔE2000 of different composite resins after immersion in the solutions (24hours and 1 month).

Immersing Solutions	Ceram X One Universal		G-aenial anterior	
	24h	1m	24h	1m
Coffee	3.78(±0.25)*	6.04(±0.41)*,**	4.19(±0.95)*	5.23(±00.62)*
Black Tea	1.49(±0.78)*	1.57(±0.67)*	1.87(±0.74)*	2.58(±0.55)*,**
Distilled water	0.79(±0.55)*	0.85(±0.33)*	0.67(±0.49)*	1.02(±0.51)*

*Mean±SD; *indicate statistical significance (p<0.05) among different staining solutions; ** indicate statistical significance (p<0.05) among different observation periods.

When examining the correlation of mean ΔE values among staining solutions and same composite resin it was found that, for coffee solution, there was statistically significant difference between ΔE values at 24 hours and one month, with ΔE (1month) being significantly higher than ΔE (24hour) for Ceram X one Universal. Whereas there was no statistically significant difference between ΔE values at 24 hours and one month found for Ceram X one-Universal specimens immersed in tea solution. Additionally, in distilled water, there was no statistically difference between ΔE values at 24 hours and one month. At 24 hours, there was statistically significant difference between ΔE values of coffee and tea, with ΔE (coffee) being significantly higher than ΔE (tea). These results were repeated at one-month observation time where also statistically significant difference was observed between ΔE values of coffee and tea, with ΔE (coffee) being significantly higher than ΔE (tea).

As far as G-aenial Anterior composite concerns, for coffee solution, there was no statistically significant difference between ΔE values at 24 hours and one month, while, in tea solution there was statistically significant difference between ΔE values at 24 hours and one month, with ΔE (1month) being significantly higher than ΔE (24hour). Additionally, specimens immersed in distilled water showed statistically significance difference between ΔE values at 24 hours and one month.

Discussion

As dental restorative materials are constantly exposed to beverages, food colourants and saliva, it is crucial to investigate their intrinsic colour stability and staining resistance, since this will compromise the restorations' imperceptibility [1, 4]. ΔE values are used for characterization of clinically perceptible changes in esthetic restorations. According to Paravina et al. ΔE_{00}=0.9 was found to be undetectable, ΔE_{00}<1.7 were found to be clinically acceptable, the 50:50 replacement point was ΔE_{00}=2.3, while $\Delta E00$=3.1 was found to be a poor match [28].

Colour stability depends on the composition of the resin matrix, dimensions of filler particles, polymerization depth and colouring agents [4]. Regarding the influence of resin matrix on colour stability, Fonseca et al. have stated that among usually used monomers, the highest degree of discolouration was recorded for BisGMA, followed by UDMA and BisEMA [29]. Previous studies have shown that silorane-based composites demonstrate lower ΔE values, after immersion in red wine, coffee or black tea, when compared to methacrylate-based resin matrixes [4, 30]. Additionally, TEGDMA containing materials appear to have less

colour stability which can be attributed to high water sorption leading to release of monomers and greater staining of the matrix [30-32]. Ceram X one Universal consists of a modified version of polysiloxane combined with polyurethane methacrylates, BisEMA and TEGDMA, while G-aenial anterior consists of a mixture of urethane dimethacrylate (UDMA) and dimethacrylate co-monomers and is BisGMA free. Colour stability of the tested materials was similar against coffee for both observation period, while Ceram X one Universal proved to be more stable after one-month immersion in black tea. These results are in accordance with previous studies and could be explained by the polysiloxane matrix of Ceram X one Universal [4, 30].

Other ingredients like photoiniators, inhibitors and comonomers could also affect the colour stability of a material, so the photoiniator system could also influence the discolouration rate of composites. Manojlovic et al. have shown that camphorquinone (CQ)/amine-based composites present higher ΔE values than Lucirin TPO-based composites [33]. In our study both materials use CQ as photoiniator, so no correlation can be made between photoiniator system and colour stability.

Water sorption is an important factor that can directly affect colour stability. Although, filler particles do not absorb water, the polymer matrix does. This means that higher proportion of resin matrix would lead to greater sorption and weaker bond between the matrix and the filler particles. As a result, higher amount of fillers and increased particle size leads to decreased colour change [1, 16, 34, 35]. Microhybrid composite seems to be more staining resistant than nanocomposite and microfilled composite [36]. In contrast to these findings, in our study, G-aenial, a hybrid composite with higher filler volume (64%) did not seem to have advanced colour stability against Ceram X one, a nanohybrid one with lower filler volume (61%). In the present study ΔE values were found to be lower than 1.7 (ΔE<1.7) for specimens immersed in water, thus this fact implies that water absorption by itself did not alter the colour of composites to a considerable extent, which is in accordance with other studies [4, 14, 37].

Microgaps located at the interface between the resin matrix and the filler are convenient pathways for colourants [16]. When a uniform distribution of the filler inside the polymer matrix is succeeded, there are less filler-depleted areas within the structure, that can be susceptible to absorption and discolouration [38]. Regarding the staining potential, coffee showed the highest potential followed by tea and distilled water for both materials. These findings are in agreement with other studies,

which mention that coffee caused the greatest discolouration followed by tea [4, 16, 18, 32]. Additionally, in the present study ΔE values for both composites immersed in coffee were greater than 3.1 (ΔE>3.1). Staining potential of tea is attributed to yellow colourants, tannic and gallic acid which are adsorbed on the superficial layer of materials and can be easily removed due to their high polarity. On the other hand, coffee contains lower polarity yellow stain molecules which cannot be removed due to their attraction and compatibility with the resin matrix [16, 27, 39].

Finally, in this study, ideal conditions were ensured concerning polymerization process, surface smoothness - flatness, that are crucial factors influencing colour stability, and thus, eliminating some of the difficulties that arise at clinical practice. Oxygen inhibited layer of the resin is removed by polishing of restorations, this procedure improves colour stability and increase surface hardness [40]. According to previous studies *in vitro* immersion of four weeks should be considered equivalent to 2.5 years of *in vivo* aging. Therefore, proportionately 24 hours of immersion *in vitro* corresponds to one month of *in vivo* aging, this is the reason why, these two periods of observation were chosen [4, 32]. In order to avoid bias, because of subjective evaluation of colour change, a spectrophotometric device was used, that allowed quantitative colour assessment and the CIEDE 2000 system, which includes three weighting functions [4, 9, 16, 18].

It is notable that when delivering a direct restoration to patients, several factors play important role on final behaviour of restorative materials over time. Apart from type of composite, shade, staining vulnerability of composite resins, the patient's dietary or social habits and compliance to oral hygiene [4]. Additionally, further *in vivo* research should be conducted in order to confirm the results of the present report.

Conclusion

Taking into consideration the limitations of this *in vitro* study, the conclusions drawn were:

i. Colour change (ΔE) at 24 hours of immersion was material independent, but solution dependent, with higher values being recorded at coffee.
ii. Colour change (ΔE) at coffee, at one month of immersion was material independent.
iii. Colour change (ΔE) at black tea, at one month of immersion was material and time dependent, where Ceram X one universal seemed to be more colour stable.
iv. Coffee had the greatest staining potential.

Acknowledgements

Conflicts of Interest

None.

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