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The Effects of Colored Pigments on the Translucency of Experimental Dental Resin Composites

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ABSTRACT

*Objective: Colored pigments are incorporated into dental resin composites to produce clinically acceptable shades for restorative materials but studies on their effects on translucency are rare. The aim of this study was to examine the effects of the addition of different colored pigments on the translucency of experimental dental resin composites. Materials and Methods: 12 types of experimental dental resin composites containing different concentrations of red and yellow iron oxide pigments were formulated and light-cured. Total and diffuse translucency as well as CIE L*a*b* values were measured and the color differences were calculated. Results: There was a statistically significant difference in the translucency values between the composites with no pigments and the composites with increasing concentrations of the pigments ($p<0.05$). The translucency decreased as the concentration of the pigments increased. However at pigment concentrations greater than 0.02%, the translucency of the composites reached a plateau and ceased to be influenced by the addition of the pigments ($p>0.05$). All color differences were in the range of 3.62-16.00 ΔE^*ab unit. Conclusions: The pigments used in this study can influence the translucency of the experimental resin composites and should be considered as an important factor by clinicians to achieve optimal esthetic restorative outcome.*

INTRODUCTION

Color and translucency of dental materials are two of the main important parameters that can significantly affect the esthetic outcomes of tooth-colored restorations.[1-4] For ideal esthetic results, it is often extremely difficult to match the shade of the restorative material to the remaining tooth structure and to the adjacent teeth. This is because of the inherent translucency of the natural enamel, which is an important optical property of the natural teeth.[5] The color and translucency of human teeth vary from one tooth or area of the mouth to another and from patient to patient.[6] Therefore, due to the increasing demand of esthetic restorations, dental restorative materials should mimic not only the color, but also the translucency of the natural teeth as closely as possible.[7]

It has been shown that different factors can influence the color and translucency of resin composites.[3,8-12] Investigated parameters include thickness, shade and background environment,[3,13,14] resin matrix composition,[15] depth of cure,[16] filler content and particle size,[17] opacifier type and concentration[18], and the initiator and filler coupling agent.[19]

However, studies on the relationship between the color and the translucency of esthetic dental resin composites are rare.[20]

Colorants and pigments are added to restorative materials to produce different shades enabling the clinicians to match the color of restorations and prostheses to the surrounding tissues.[21]

Colored pigments used in dental resin composites include a combination of white titanium oxide, red iron oxide, yellow iron oxide and black iron oxide with different proportions. These pigments may interact with each other and with the components of the composite paste affecting the optical properties of the composite material. Studies evaluating the influence of these pigments on the translucency of resin composites are rare and the influence of pigments on light transmission has been reported in only one study very recently.[22]

Therefore, the aim of this study was to investigate the effects of the addition of different individual colored pigments and their combinations on the translucency of the experimental dental resin composites.

MATERIALS AND METHODS

COMPOSITE RESIN FORMULATION

All the materials used in this study for fabrication of the experimental composites were supplied by DENTSPLY (Konstanz, Germany).

The resin matrix was prepared by mixing the following ingredients (in wt.%): urethane dimethacrylate (UDMA) (99.22%), camphorquinone (CQ) (0.3%), dimethylaminobenzoic acid ethyl ester (DMABE) (0.3%), 3,5-di-tert-butyl-4-hydroxytoluene (BHT) (0.12%) and 2-hydroxy-4-methoxybenzophenone (HMBP) (0.06%).

The experimental resin composites were produced by mixing 25wt.% of resin matrix with 75 wt.% of silane-treated barium-silicate filler containing different concentrations of red and yellow iron oxide pigments as shown in Table 1. Composite 1 was the control group and contained no pigments. Composite 2 to 5 only had red pigments. Composites 6 to 9 only had yellow pigments and composites 10 to 12 had both red and yellow pigments in different quantities. For every composition, the filler content was kept constant at 75%, so that only the effect of the changing concentrations of the pigments would be studied.

SPECIMEN FABRICATION

The ingredients were measured for the desired weight using an analytical balance (Mettler AJ100, Greifensee, Switzerland) and then were mixed by hand in small flexible plastic containers. Once mixed to a homogeneous paste, the experimental resin was ready to be placed into the molds.

A polycarbonate sheet of 1.5 mm thickness, containing six holes of 15.5 mm diameter, was made to act as mould for the specimens. Each group of unpolymerized resin composite specimens was packed into the six moulds over a glass plate using a condenser, making sure no bubbles were created. Another glass plate was placed over the polycarbonate sheet and firm pressure was applied for twenty seconds. The specimens were then light-cured from both sides in three different locations for a total of 90 seconds. The light source unit (QHL 75, Dentsply) had an irradiance setting of 450mW/cm^2.

Of the six polymerized specimens, three were chosen based on homogeneity and lack of porosities. The other three were discarded. A total of thirty-six specimens were selected for the study (N=36).

The specimens were ground from both sides using a silicon carbide grinding paper (Buehler-Met® II, Buehler UK, Coventry) P400 to the thickness of 1.3 mm, and subsequently polished with a P1200 to the thickness of 1 mm (±0.05 mm) for a smooth finish. This was carried out on a grinder-polisher machine (Buehler Metaserv, Buehler UK) rotating at 200 rpm speed. A micrometer was used to check thickness of the specimens in five different locations (one at the centre and four at the edges). A bright light source was used to check for porosities. Specimens that showed inappropriate thickness and/or porosities were discarded and replaced.

Each specimen was then rinsed with water, dried and stored with the other two specimens of the same group in a dry environment in a self-sealing small polythene bag.

Table 1. Pigment type and content of different experimental dental resin composites

Experimental resin composite	Red iron oxide pigment (%)	Yellow iron oxide pigment (%)
1 (control group)	0	0
2	0.02	0
3	0.04	0
4	0.06	0
5	0.08	0
6	0	0.01
7	0	0.02
8	0	0.03
9	0	0.04
10	0.02	0.03
11	0.04	0.02
12	0.06	0.01

MEASUREMENT OF OPTICAL PROPERTIES

Optical properties data were collected using a computer-controlled spectrophotometer (Lambda 2, PerkinElmer, Massachusetts, USA) with integrating sphere. Transmittance (total, diffuse and total direct) was measured in the wave length range of 380-700 nm under standard illuminant D65 at 1 nm intervals. Color coordinates, L* (lightness), a* (red-green chromaticity index), and b* (yellow-blue chromaticity index) were determined from the total transmittance data using Pecol color software (PerkinElmer, USA).

For Total Transmittance and Diffuse Transmission, measurements were taken for every wave length from 380 nm to 700 nm, resulting in 321 readings. For Total Transmittance measurement, a specimen was placed in the transmission port (entry port) of the spectrophotometer and a white reference material was placed in the reflectance port (*Figure 1*).

For Diffuse Transmission, a light trap needs to exist in the reflectance port. The light trap absorbs the direct transmission, and therefore only scattered light is measured. A light trap can be either a black background or an open port. In this study, an open port was chosen as a light trap.

For direct transmittance, the values of total transmittance were subtracted from diffuse transmittance, to measure light passing through the samples without scattering.

Color measurements were taken using CIE Lab values in total transmittance mode. Color difference (ΔE*) was measured using the following equation:

$$\Delta E^* ab = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{0.5}$$

DATA ANALYSIS

Statistical analysis of the data was carried out by one-way ANOVA followed by Tukey's test with a confidence interval (CI) of 95% using Minitab statistical analysis software.

Regression analysis was used to plot the line of the best fit for the CIE L*a*b* values.

RESULTS

The mean total, diffuse and direct transmittance of the composite formulations with different concentrations of red pigment, yellow pigment and mixture of red and yellow pigments are shown in Figures 2, 3 and 4 respectively.

Statistical analysis by one-way ANOVA followed by Tukey's test showed that there was a statistically significant difference ($p<0.05$) in the translucency values between the composites with no pigments and the composites with increasing concentrations of the pigments. The translucency decreased as the concentration of the pigments increased. However at pigment concentrations greater than 0.02% as shown in the Figures 2 and 3, the translucency of the composites reached a plateau and ceased to be influenced by the addition of the pigments. The yellow and red pigments showed a similar pattern.

Figure 4 shows the translucency of the composite resins with mixed red and yellow pigments, where the concentration of the red pigment is gradually increased and the concentration of the yellow pigment is gradually decreased. All mixture combinations reduced the translucency of the composite resins except for the mixture of 0.06% red and 0.01% yellow pigment.

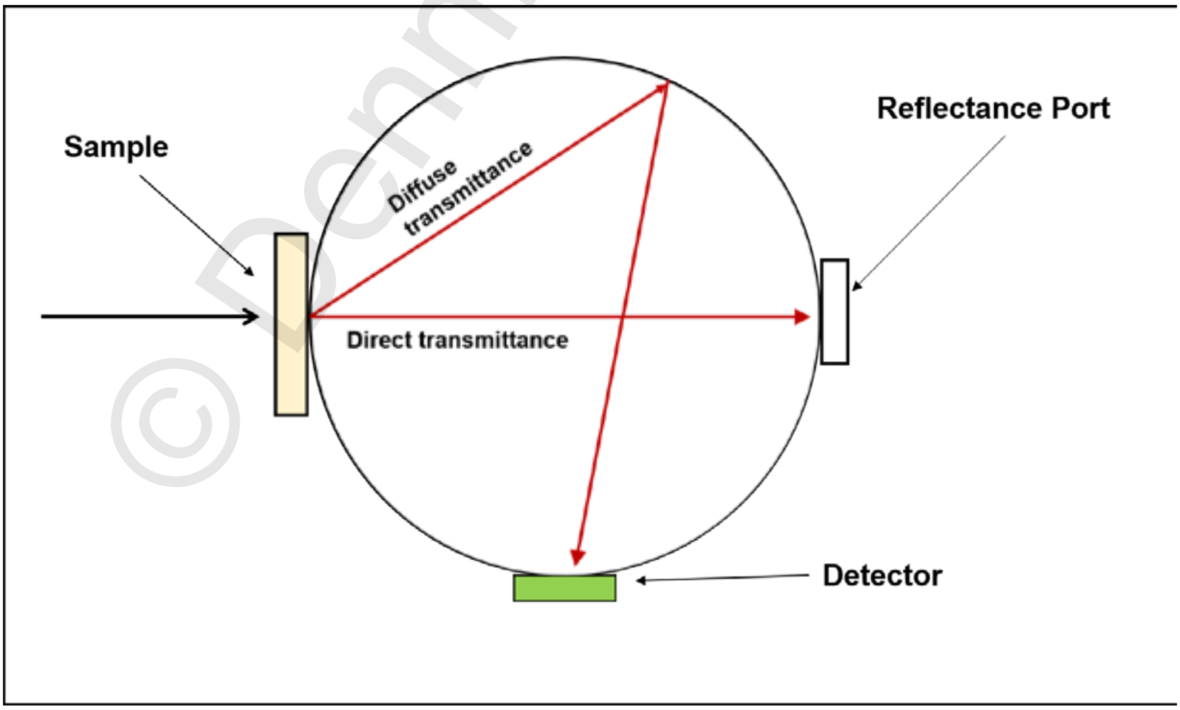

Figure 1: Schematic diagram of the mechanism of light transmittance detection by spectrophotometer

Regression analysis showed that there was a linear correlation between the concentrations of the pure red, pure yellow pigments and total translucency.

CIE L*a*b* results showed that for the yellow pigment, the lightness decreased with an increase in the concentration whereas for the red pigment the yellowness of the sample increased. The a* value for the yellow pigment decreased and for the red pigment increased. The b* value for both yellow and red pigment increased with the increase in the concentrations of the pigments. The mixed pigments showed variations in the lightness.

Measurement of the color differences (ΔE*ab) of resin composites with different concentrations of pigments showed all color differences were in the range of 3.62–16.00 ΔE*ab unit.

DISCUSSION

Resin composites have become the most popular restorative materials in recent years due to several reasons including 1) increased demand for aesthetic restorations, 2) environmental concerns and health concerns of mercury, 3) suitable mechanical properties and 4) as alternative to amalgams.[23] Whilst resin composites are deemed to have suitable optical properties and the ability to be placed directly, these may not arguably be the main reasons for its popularity their optical properties and an increasing demand for a directly placed esthetic restorative material. Several factors influence the esthetics of a restorative material, such as color, translucency and surface texture.[24] Many studies have investigated the color of composite resins and the studies on the translucency of the composite resins have mainly focused on parameters such as filler type, size and content, resin matrix refractive index and the monomer type.[3,8-12]

The present study was the first study to evaluate the effect of additions of colored pigments with different concentrations on the translucency of resin composites. The individual effects of the red and yellow pigments, as well as the combined effects of these pigments were investigated.

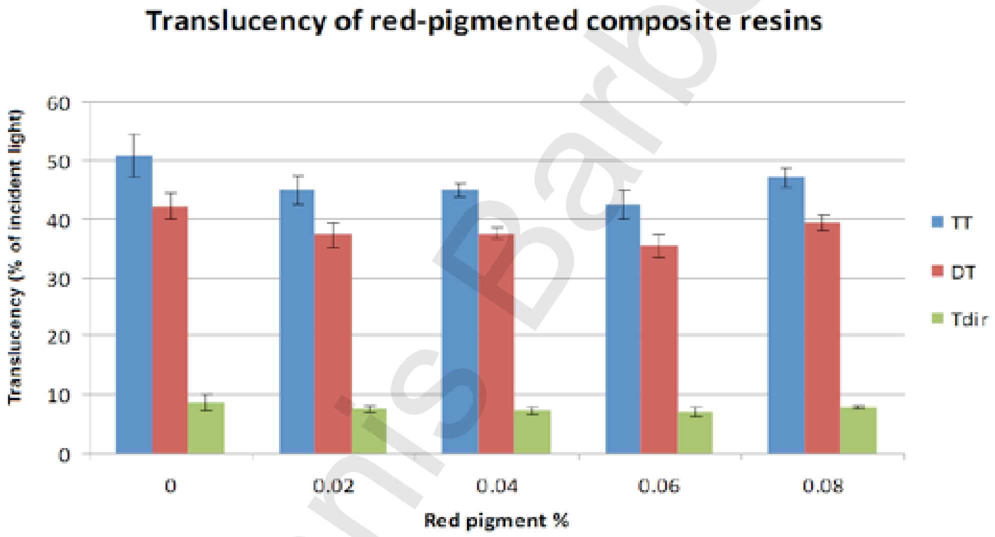

Figure 2: Translucency of composite resins with increasing concentrations of the red-pigment. The translucency values decrease and reach a plateau with an increase in the pigment concentration.

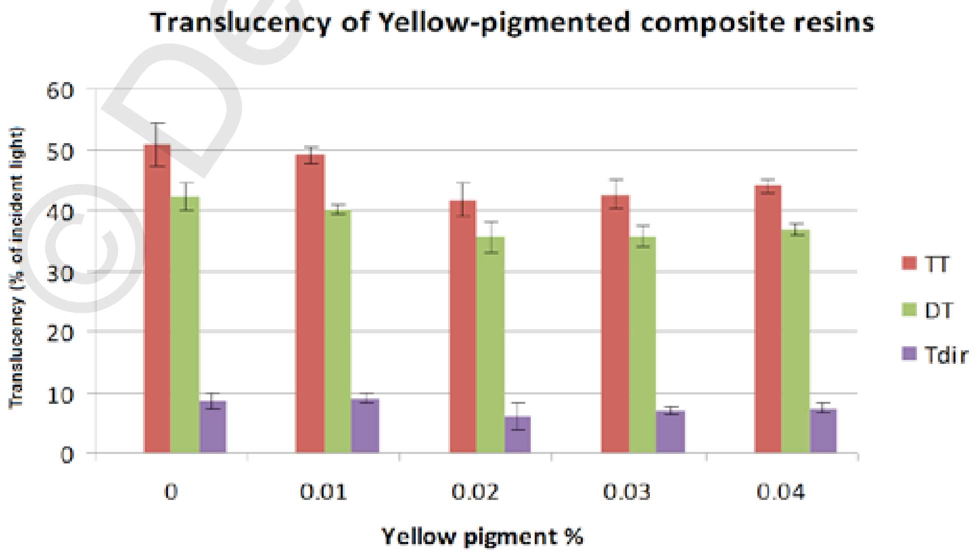

Figure 3: Translucency of composite resins with increasing concentrations of the yellow-pigment. The translucency values of the composite resin decrease with an increase in the concentration of the yellow pigment.

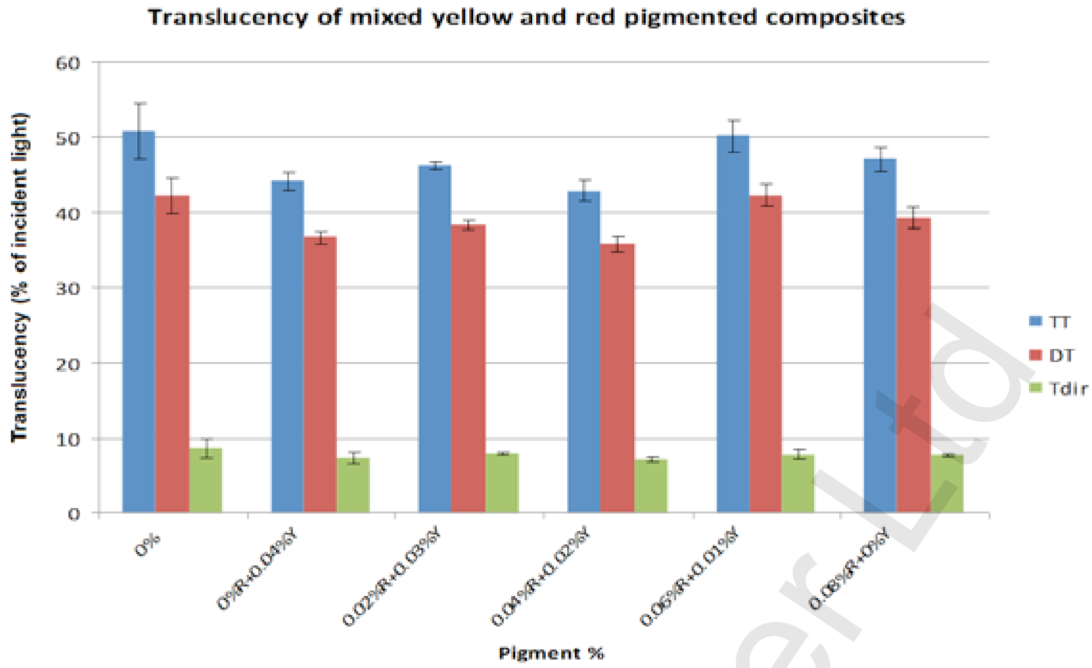

Figure 4: Translucency of composite resins with increasing concentrations of the mixed-red and yellow pigments.

Experimental resin composites were formulated in order to eliminate the variables such as different additives, manufacturing conditions, monomer type and filler concentrations that are used in commercially available products. All composite samples were made using the UDMA monomer and the filler content was kept constant at 75% for all specimens. UDMA and BisGMA are widely used as the base monomers in the formation of polymeric matrices of the dental composites.[25] Unlike, BisGMA, UDMA does not need to be diluted with a viscosity controller and facilitates consistent composite material fabrication.

The most commonly used pigments in commercially available composite resins are metal oxide pigments, such as red ferric oxide Fe_2O_3 or yellow ferric trihydroxide $Fe(OH)_3$,[26] which were generously supplied by DENTSPLY for this study. The pigment concentrations used in this study were based on the recommendations from the manufacturers of dental resin composites.

Porosity within the composite can cause light scattering and interfere with the results and thus it is important to eliminate any porosity and air bubbles from the samples. Careful packing of the uncured resin in the mould and then pressing it between two glass slabs with hand pressure during curing was a useful technique to minimise the porosity and produce smooth samples with even thickness as suggested by some other studies.[27]

The light transmittance through the composite resin was measured using a Spectrophotometer (Elmer Lambda 2 UV/VIS spectrophotometer) with an integrating sphere. Spectrophotometers are widely used to study the optical properties of restorative materials such as ceramics and composites because they provide an accurate and reproducible method of light transmission measurement.[28] Advanced systems that incorporate the latest spectrophotometric and computer technology, have significantly increased the capabilities for measuring and controlling color in many fields.[28]

From the results of this study it could be seen that there was a statistically significant difference in the translucency of the resin composites after the addition of the pigments. Following the addition of the red pigment, the TT and DT values first decreased up to a concentration of pigment of 0.06% and then started to increase again at 0.08% pigment concentration. Similarly for the yellow pigments, a similar pattern was observed, i.e. a decrease in TT and DT up to 0.02% pigment content and an increase from 0.04% concentration onwards. It seems from the line graphs that for the yellow and red pigments, the addition of pigments affected all the wavelengths of the visible spectrum.

The pigment particles act as scattering centres, similar to opacifiers, thereby scattering more light as their concentrations increase, causing a decrease in the translucency of the resin composites tested in this study. This is consistent with previous studies which have shown that darker shades of resin composites which have more pigment contents are less translucent.[29]

It was observed that when the yellow and red pigments were mixed together, the translucency reduced initially as expected but started increasing again with the increasing concentration of red pigment and a decreased concentration of yellow pigment. It seems that the addition of red and yellow pigment does affect the translucency to some extent, but the TT and DT values increase when a mixture of 0.06% red pigment and 0.01% yellow pigment is used.

To understand the above results better, line graphs were made between the individual wave lengths starting from 380 to 700 for the translucency of the mixed, red and yellow pigment concentrations. It appeared from these graphs that there was a rotation around one point of wavelength possibly because at that wavelength more light was absorbed by the pigments hence decreasing the translucency. With the addition of the pigment relatively more light in the red and yellow wave length range is allowed to pass through compared with the blue/green range. Thus while the overall translucency is only slightly affected, there is a large change in the color.

From the CIE L*a*b* values of the yellow pigment it could be seen that the lightness decreased with an increase in the concentration of the yellow pigment. The a* value, which is a measure of the red-green content, decreased as predicted. The b* value, which is a measure of the yellow-blue content, increased with the increase in the yellow concentration. A similar pattern for the red pigment could be seen with a* values increasing with the increase in the concentration of the red pigment. The b* value also increased to some extent with an increase in the red pigment.

The CIE L*a*b* values for the mixed pigment showed a fluctuation in the lightness and there was a linear relationship between the mixed pigment concentration and the a* value. Similarly it could be seen that, the a* value also had a linear relationship, but it decreased as the concentrations of the mixed pigments changed. The mixed pigments ideally should show an effect between the patterns shown by the red and the yellow pigment separately. To understand this better all the L*a*b* values of the red, the yellow and the mixed pigments were plotted on a CIE L*a*b* space.

The scatter graph showed that when yellow pigment was added, there was a large yellow shift in the CIE L*a*b* space. But when red-pigment was added there was a red as well as yellow shift. This showed that with the addition of red-pigment, the yellowness of the sample also increased.

The red shaded area in the figure below showed large shift towards the Y as well as the X-axis i.e. a shift towards red and yellow space. Similarly the green shaded area indicates that there was only yellow shift after the addition of the yellow pigment. Following the addition of the red and yellow pigments both, the values for the mixed pigment stayed somewhere between the a* and b* as shown in the Figure 5.

Color differences (ΔE*ab) of resin composites with different concentrations of pigments were also determined, which showed color differences were in the range of 3.62–16.00 ΔE*ab unit. All the combinations showed ΔE*ab values higher than 1 which is considered to be the threshold level for a color change between two materials to be perceptible.[30] If the ΔE is taken as ΔE> 3.3 which is indicated as the clinically perceptible color difference according to many studies, all the samples showed a perceptible difference. The minimum difference was

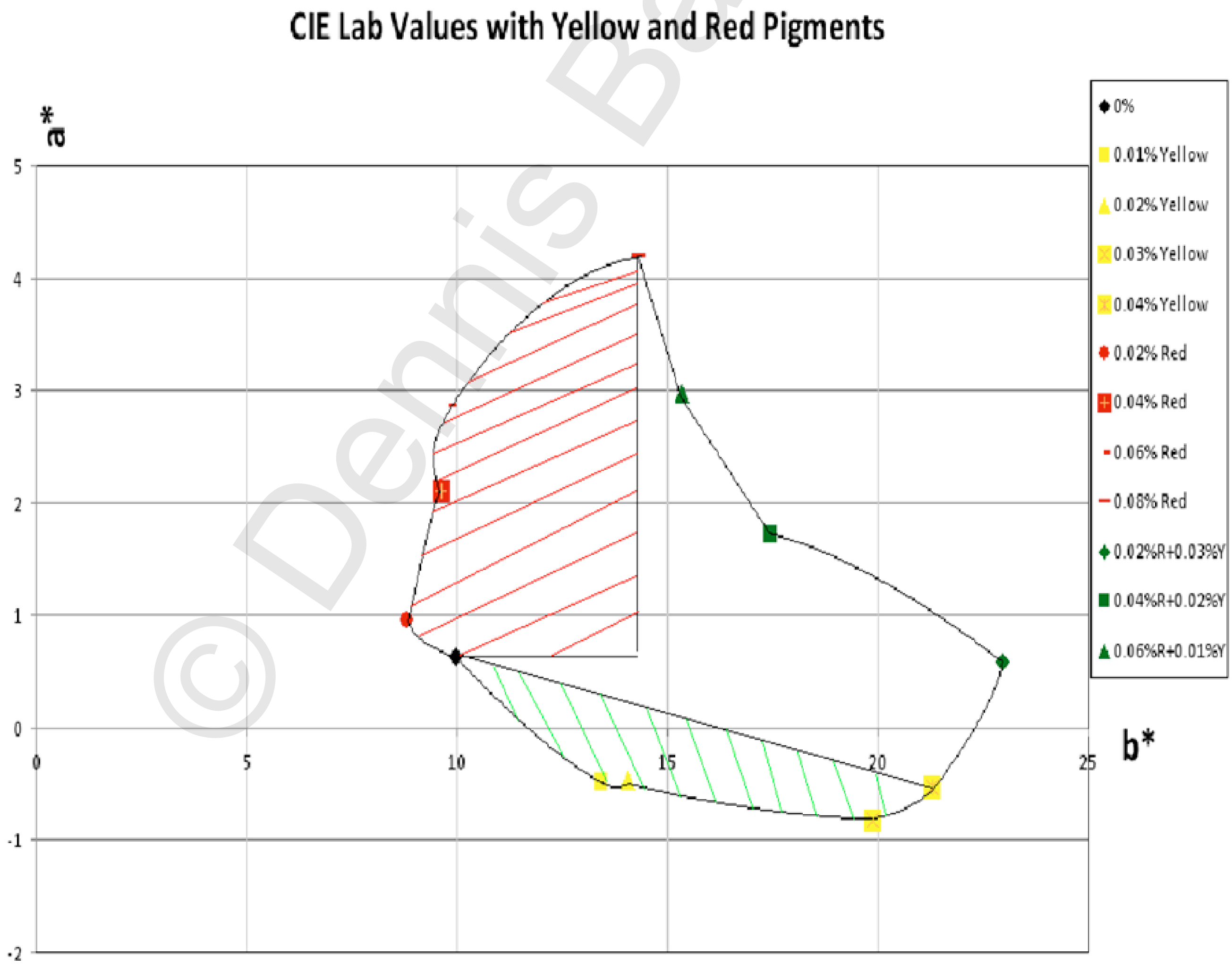

Figure 5: Scatter Graph of CIE Lab values for the experimental composites with different pigment contents

3.62ΔE* units which was the difference between the control group and 0.04% yellow pigmented sample. The relevance of this finding is that these differences are clinically perceivable as it can be seen that even minor concentrations of pigments can bring about a highly distinguishable change in the color.

The results of this study indicate that the addition of the red and yellow colored pigments to the resin composites affects their translucency to some extent. These findings have not been reported previously and are consistent with the suggestions by only one previous study investigating the relationship between the color and translucency of commercially available multi-shade resin composites, which showed the translucency of the composites decreased significantly towards the darker shades.[20]

CONCLUSIONS

From the results of this study it can be concluded that the addition of red and yellow iron oxide pigments significantly reduces the translucency of experimental dental resin composites. However, with further increases in the concentration of the pigments, the translucency stops being affected and acquires a constant value.

The addition of yellow pigment causes a large change in b* value, whilst L and a* only show small changes. Thus the predominant shift in color is towards the yellow. The addition of red pigment causes both a change in a* and b*, thus producing a shift towards red as well as yellow.

Since the pigments used in this study can influence the translucency of the experimental resin composites, it should be considered as an important factor by clinicians to achieve optimal esthetic restorative outcome.

REFERENCES

1. Villarroel M, Fahl N, De Sousa AM, De Oliveira OB, Jr. Direct esthetic restorations based on translucency and opacity of composite resins. *J Esthet Rest Dent*. 2011;**23**:73-87.
2. Brodbelt RH, O'Brien WJ, Fan PL. Translucency of dental porcelains. *J Dent Res*. 1980;**59**:70-75.
3. Miyagawa Y, Powers JM, O'Brien WJ. Optical properties of direct restorative materials. *J Dent Res*. 1981;**60**:890-894.
4. ten Bosch JJ, Coops JC. Tooth color and reflectance as related to light scattering and enamel hardness. *J Dent Res*. 1995;**74**:374-380.
5. Lee YK. Influence of scattering/absorption characteristics on the color of resin composites. *Dent Mater*. 2007;**23**:124-131.
6. Powers JM. Restorative dental materials. 12th ed. St. Louis: Mosby; 2006.
7. Kim JH, Lee YK, Powers JM. Influence of a series of organic and chemical substances on the translucency of resin composites. *J Biomed Mater Res B Appl Biomater*. 2006;**77**:21-27.
8. Lim YK, Lee YK, Lim BS, Rhee SH, Yang HC. Influence of filler distribution on the color parameters of experimental resin composites. *Dent Mater*. 2008;**24**:67-73.
9. Leloup G, Holvoet PE, Bebelman S, Devaux J. Raman scattering determination of the depth of cure of light-activated composites: influence of different clinically relevant parameters. *J Oral Rehabil*. 2002;**29**:510-515.
10. Emami N, Sjodahl M, Soderholm KJ. How filler properties, filler fraction, sample thickness and light source affect light attenuation in particulate filled resin composites. *Dent Mater*. 2005;**21**:721-730.
11. dos Santos GB, Alto RV, Filho HR, da Silva EM, Fellows CE. Light transmission on dental resin composites. *Dent Mater*. 2008;**24**:571-576.
12. Masotti AS, Onofrio AB, Conceicao EN, Spohr AM. UV-vis spectrophotometric direct transmittance analysis of composite resins. *Dent Mater*. 2007;**23**:724-730.
13. Powers JM, Dennison JB, Lepeak PJ. Parameters that affect the color of direct restorative resins. *J Dent Res*. 1978;**57**:876-880.
14. Arikawa H, Fujii K, Kanie T, Inoue K. Light transmittance characteristics of light-cured composite resins. *Dent Mater*. 1998;**14**:405-411.
15. Azzopardi N, Moharamzadeh K, Wood DJ, Martin N, van Noort R. Effect of resin matrix composition on the translucency of experimental dental composite resins. *Dent Mater*. 2009;**25**:1564-1568.
16. Taira M, Okazaki M, Takahashi J. Studies on optical properties of two commercial visible-light-cured composite resins by diffuse reflectance measurements. *J Oral Rehabil*. 1999;**26**:329-337.
17. Yeh CL, Miyagawa Y, Powers JM. Optical properties of composites of selected shades. *J Dent Res*. 1982;**61**:797-801.
18. Haas K, Azhar G, Wood DJ, Moharamzadeh K, van Noort R. The effects of different opacifiers on the translucency of experimental dental composite resins. *Dent Mater*. 2017;**33**:e310-e316.
19. Johnston WM, Reisbick MH. Color and translucency changes during and after curing of esthetic restorative materials. *Dent Mater*. 1997;**13**:89-97.
20. Naeimi Akbar H, Moharamzadeh K, Wood DJ, Van Noort R. Relationship between Color and Translucency of Multishaded Dental Composite Resins. *Int J Dent*. 2012;**2012**:708032.
21. Johnston WM, Ma T, Kienle BH. Translucency parameter of colorants for maxillofacial prostheses. *Int J prostho*. 1995;**8**:79-86.
22. Palin WM, Leprince JG, Hadis MA. Shining a light on high volume photocurable materials. *Dent Mater*. 2018;**34**:695-710.
23. Mulligan S, Kakonyi G, Moharamzadeh K, Thornton SF, Martin N. The environmental impact of dental amalgam and resin-based composite materials. *Brit Dent J*. 2018;**224**:542-548.
24. Joiner A. Tooth colour: a review of the literature. *J Dent*. 2004;**32** Suppl 1:3-12.
25. Dhuru VB. Contemporary dental materials. New York: Oxford University press; 2004.
26. Simone Klapdohr NM. Invited Review: new inorganic components for Dental filling composites. *Chemical Monthly*. 2005;**136**:21-45.
27. Nakamura T, Tanaka H, Kawamura Y, Wakabayashi K. Translucency of glass-fibre-reinforced composite materials. *J Oral Rehabil*. 2004;**31**:817-821.
28. Beering M. Techniques for measuring color. *Metal Finish*. 2000;**98**:565-570.
29. Yu B, Lee YK. Influence of color parameters of resin composites on their translucency. *Dent Mater*. 2008;**24**:1236-1242.
30. Johnston WM. Color measurement in dentistry. *J Dent*. 2009;**37** Suppl 1:e2-6.

