Color adjustment potential of single-shade resin composite to various-shade human teeth: Effect of structural color phenomenon

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This study evaluated the effect of the structural color phenomenon in resin composites (RCs) on the color adjustment of restorations by investigating their color reproduction performance in human incisors of various shade. Cervical cavities were filled with a single-shade RC with 260 nm spherical fillers (Omnichroma (OMN)), conventional A2-shade RCs (Estelite Σ Quick or Clearfil AP-X), or experimental RCs with 5–50 nm fumed silica fillers (R1) and 100 nm spherical fillers (R2). Color parameters ($L^*a^*b^*$) were measured using a CIE XYZ camera along the centerline of the restorations, and the color difference ($\Delta E_a^*$) between corresponding areas of intact and restored teeth was calculated. Additionally, the reflectance spectra of OMN, R1, and R2 were investigated. OMN exhibited significantly lower $\Delta E_a^*$ than other tested RCs ($p<0.05$) and its reflection spectrum ranged from blue to red, while a blue peak was observed with R1 and R2, indicating a higher color adjustment potential of OMN.

Keywords: Structural color, Color adjustment, Resin composite, Human teeth, Filler size

INTRODUCTION

Resin composites are now broadly used as esthetic restorative materials for anterior and posterior teeth because of their improved physical and mechanical properties, as well as cosmetic appearance through the developments in filler technology. However, matching the color of the resin composite with the surrounding tooth remains a difficulty, because tooth color is affected by several factors, such as the type of tooth, site, and age. Therefore, color-matching techniques are required, such as shade selection and multi-layered filling approaches using composite materials of various shades that have been adjusted with pigments or dyes. Nonetheless, it is well known that the perceived color difference between the resin composite and the surrounding tooth is less than that envisaged from observing the colors in isolation due to mutual color blending resulting from the discrimination of wavelengths by the interaction of incident light with nanostructures like thin-films, diffraction gratings, or photonic crystals. As structural colors are the result of fundamental optical processes of diffraction, interference, and scattering, they do not fade, as opposed to conventional colors which originate from light absorption by pigments. Depending on their optical properties, structural colors are classified as iridescent or non-iridescent. Iridescent structural colors appear to gradually change depending on the angle of illumination or the orientation of the viewer, while non-iridescent structural colors, which are said to occur in colloidal amorphous aggregates, are independent of the angle of observation and appear consistent.

The application research of structural colors in industries such as textile and automotive is ongoing. Among the methods of manufacturing artificial structural colors, bottom-up methods based on assembling colloidal nanoparticles are considered to be the most promising in adjusting the wavelength range of the structural color, because it is assumed that the size of the minute structures enhances a specific color tone. In the case of resin composites, the filler particle size and shape

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are likely to be an important factor in achieving the phenomenon of structural color as ambient light passes through the composite material. The manufacturer of OMN claims that this phenomenon is induced by using spherical filler particles homogenized to 260 nm in size by the “sol-gel method”, which has been used to obtain uniform filler particles with a defined reflectivity. A recent study reported that resin composites containing the 260 nm spherical fillers exhibited structural color and that their color adaptation to denture teeth of various shades was excellent, but their color adjustment potential in various-shade human teeth remains unclear.

Therefore, the purpose of this study was to evaluate the effect of the structural color phenomenon of OMN on the color adjustment potential using human incisors of various shades, by investigating whether composite restorations in cervical cavities could reproduce the original color of the tooth. The color reproduction performance of OMN was compared to two commercial multi-shade composites, and was also compared to two experimental composites with different particle size, which had similar transparency and light transmission properties but not structural color, in order to investigate the effect of the structural color phenomenon on the color adjustment of the restoration. The null hypothesis tested was that the structural color phenomenon does not affect the color adjustment potential of the tested composites in human teeth of various shades.

**MATERIALS AND METHODS**

**Specimen preparation**

Thirty extracted human central incisors without endodontic treatment or restorations, were collected and stored in water at 4°C. The research was approved by the Human Research Ethics Committee of Tokyo Medical and Dental University (ethical protocol No. 2013-022) and informed consent was obtained from the participants before the teeth were extracted. The shade distribution (shade A1-4; B1-4; C2-4; D2,4) of the teeth was determined using a dental spectrophotometer (Crystaleye; OLYMPUS, Tokyo, Japan). Color images of the teeth were captured in a black box with 100% relative humidity using a CIE XYZ digital camera (RC500, PaPaLaB, Shizuoka, Japan) from a distance of 20 cm. The advantage of the CIE XYZ color space is that it avoids the negative sensitivity of the RGB isochromatic function. Additionally, the sensitivity of the RC500 camera is equivalent to that of the human eye, thus faithfully reproducing the human perception of colors. The exposure duration was set to 0.2 s and the shutter speed to 1/1000–1/15 s. The scene was spotted with a D65 standard illuminant with 45/0-degrees geometry on both sides. After the images of the intact tooth surfaces were taken, a cylindrical cavity (diameter: 4.0 mm, depth 2.0 mm) was prepared on the labial side at the cementoenamel junction using a cylindrical diamond bar (#148, ISO 032, Shofu, Tokyo, Japan) with a 1:5 electric handpiece.

**Color measurement of teeth restored with commercial resin composites**

Three commercially available resin composites, Omnichroma (OMN; Tokuyama Dental), Estelite Σ Quick (A2 shade) (ESQ; Tokuyama Dental), and Clearfil AP-X (A2 shade) (APX; Kuraray Noritake Dental, Tokyo, Japan) were used in this study (Table 1). After applying

### Table 1  Resin composite materials used in this study

| Material (manufacturer) | Composition | Filler type (batch number) |
|-------------------------|-------------|----------------------------|
| Omnichroma (Tokuyama Dental, Tokyo, Japan) | Filler: 79 wt% uniform sized supra-nano spherical filler (SiO₂-ZrO₂ 260 nm), round-shaped composite filler (containing 260 nm spherical SiO₂-ZrO₂) | Nanofilled (17J23) |
| Clearfil AP-X (A2) (Kuraray Noritake Dental, Tokyo, Japan) | Filler: 85 wt% silanated barium glass filler of irregular shape (700 nm), silanated silica filler (100–1,500 nm) | Microhybrid (380094) |
| Estelite Σ Quick (A2) (Tokuyama Dental) | Filler: 82 wt% uniform sized supra-nano spherical filler (SiO₂-ZrO₂, SiO₂-TiO₂ 100–300 nm (average 200 nm), round-shaped composite filler (100–300 nm (average 200 nm), spherical SiO₂-ZrO₂) | Nanofilled (J0662) |
| ECM-001R1 (Tokuyama Dental) | Filler: 19 wt% fumed silica (5–50 nm (average primary particle size 15 nm)) | Nanofilled (18D171) |
| ECM-001R2 (Tokuyama Dental) | Filler: 60 wt% uniform sized supra-nano spherical filler (SiO₂-ZrO₂ 100 nm) | Nanofilled (18E311) |

TEGDMA, triethylene glycol dimethacrylate; UDMA, urethane dimethacrylate; Bis-GMA, bisphenol-A-glycidyl methacrylate.
a self-cure self-etch adhesive (Bondmeter Lightless, Tokuyama Dental) to the cavities according to the manufacturer's instruction, one resin composite (OMN, APX, ESQ) was randomly selected and placed into the cavity using a resin filling instrument (TMDU type3 #3, YDM, Tokyo, Japan) and irradiated with a light-curing unit (Pencure 2000, Morita, Tokyo, Japan) at an intensity of 1,000 mW/cm² for 40 s. After storage in 100% relative humidity at 37°C for 24 h, the specimens were polished waterproof silicon carbide paper under running waters and stored in 100% relative humidity and 37°C for 24 h. The color of the resin composite discs over white and black backgrounds was measured using the RC500 camera. The translucency parameter (TP) was obtained by calculating the color difference of the specimen over a black and white background using the following formula:

\[ TP = [(L_B^* - L_W^*)^2 + (a_B^* - a_W^*)^2 + (b_B^* - b_W^*)]^1/2. \]

Subscript B refers to the color coordinates over a black background, while subscript W refers to those over a white background.

**Measurement of the spectral reflectance of the experimental resin composites and Omnicroma**

Three additional discs were prepared of each resin composite (OMN, R1, R2) as described above. The spectral reflectance of the resin discs was determined using the TC-1800 spectrophotometer (Tokyo Denshoku, Tokyo, Japan) over a black background. Each resin disc was measured three times, and the mean value for each wavelength was taken as its spectral reflectance.

**Statistical analysis**

The color difference data were analyzed by the Wilcoxon signed-rank test, while the TP data of the resin composite discs were analyzed using one-way ANOVA and t-tests with Bonferroni correction. All statistical procedures were performed at a confidence level of 95% using the Statistical Package for the Medical Science (IBM SPSS Statistics 20, IBM, Chicago, IL, USA).

**RESULT**

**Color of teeth restored with OMN, APX, and ESQ**

The \( \Delta L^* \), \( \Delta h^* \) \( \Delta C^* \), and \( \Delta E_{00} \) values of OMN, APX, and ESQ are shown in Figs. 1–4. OMN exhibited the lowest \( \Delta L^* \), significantly different from ESQ and APX \( (p<0.05) \). Moreover, the \( \Delta L^* \) of ESQ was a significantly lower compared to APX \( (p<0.05) \). The lowest \( \Delta h^* \) value was measured with OMN, followed by APX and ESQ whose \( \Delta h^* \) values were significantly higher than OMN \( (p<0.05) \). There was no significant difference in \( \Delta h^* \) between APX and ESQ \( (p>0.05) \). The \( \Delta C^* \) values of the three commercial composites did not differ significantly \( (p>0.05) \). OMN also exhibited the significantly lowest \( \Delta E_{00} \) value \( (p<0.05) \), followed by ESQ and APX.

**Color of teeth restored with OMN, R1, and R2**

The \( \Delta L^* \), \( \Delta C^* \), \( \Delta h^* \), and \( \Delta E_{00} \) values of OMN, R1, and R2 are given in Figs. 5–8. OMN exhibited the lowest \( \Delta L^* \) and \( \Delta h^* \) values, followed by R1 and R2 whose values were significantly higher compared to OMN \( (p<0.05) \). The significantly lowest \( \Delta C^* \) and \( \Delta E_{00} \) values were also obtained with OMN \( (p<0.05) \), followed by R2 and R1 which differed significantly from each other in both the parameters \( (p<0.05) \).

**Translucency parameter**

The TPs of the resin composites are presented in Table...
Fig. 1 $\Delta L^*$ values of the commercial resin composites Omnichroma (OMN), Clearfil AP-X (APX), and Estelite $\Sigma$ Quick (ESQ). The lines indicate significant differences between the materials ($p<0.05$, $n=30$).

Fig. 2 $\Delta C^*$ values of the commercial resin composites Omnichroma (OMN), Clearfil AP-X (APX), and Estelite $\Sigma$ Quick (ESQ). There were no significant differences between the materials ($n=30$).

Fig. 3 $\Delta h^*$ values of the commercial resin composites Omnichroma (OMN), Clearfil AP-X (APX), and Estelite $\Sigma$ Quick (ESQ). The lines indicate significant differences between the materials ($p<0.05$, $n=30$).

Fig. 4 $\Delta E_{00}$ values of the commercial resin composites Omnichroma (OMN), Clearfil AP-X (APX), and Estelite $\Sigma$ Quick (ESQ). The lines indicate significant differences between the materials ($p<0.05$, $n=30$).

Fig. 5 $\Delta L^*$ values of the commercial resin composite Omnichroma (OMN) and the experimental resin composites R1 and R2. The lines indicate significant differences between the materials ($p<0.05$, $n=40$).

Fig. 6 $\Delta C^*$ values of the commercial resin composite Omnichroma (OMN) and the experimental resin composites R1 and R2. The lines indicate significant differences between the materials ($p<0.05$, $n=40$).
Fig. 7 \( \Delta h^* \) values of the commercial resin composite Omnichroma (OMN) and the experimental resin composites R1 and R2. The lines indicate significant differences between the materials \((p<0.05, n=40)\).

Fig. 8 \( \Delta E_{00} \) values of the commercial resin composite Omnichroma (OMN) and the experimental resin composites R1 and R2. The lines indicate significant differences between the materials \((p<0.05, n=40)\).

Table 2 Translucency parameter (TP) of the commercial resin composites Omnichroma (OMN), Clearfil AP-X (APX), Estelite Sigma Quick (ESQ), ECM-001R1 (R1) and ECM-001R2 (R2) at 1 mm thickness

| Material     | APX       | ESQ       | OMN       | R1        | R2        |
|--------------|-----------|-----------|-----------|-----------|-----------|
| TP value     | 24.2 (0.79) \( ^A \) | 22.6 (0.73) \( ^B \) | 24.1 (0.29) \( ^A \) | 24.3 (0.33) \( ^A \) | 24.4 (0.67) \( ^A \) |

Mean (SD), \( n=5 \).
The TP data were statistically analysed using one-way ANOVA and t-tests with Bonferroni correction. The same superscript uppercase letters indicate no significant difference between groups \((p>0.05)\).

2. The TP values of APX, OMN, R1, and R2 were significantly higher than ESQ \((p<0.05)\), while there was no considerable difference between APX, OMN, R1, and R2 \((p>0.05)\).

Spectral reflectance of OMN, R1, and R2

The reflectance spectra of OMN, R1, and R2 are shown in Fig. 9. The spectral reflectance of R1 and R2 peaked at approximately 450 nm (blue range), while the intensity gradually decreased in the longer wavelength region. Although, OMN exhibited an increasing spectral reflectance at approximately 450 nm as well, its spectral reflectance remained high at longer wavelengths, until ~750 nm.

DISCUSSION

The color developments are typically classified as either structural or pigmentary\(^{30}\). The mechanism of pigmentsary colors produced by the energy consumption of light entails that pigments selectively absorb some wavelengths of light, while allowing others to be reflected\(^{31}\). Generally, light-cured resin composites are prepared with the various shades by adding pigments to correspond to various tooth colors. In this study, ESQ and APX adjusted to the A2 shade by pigments were used to fill cavities prepared in teeth of various shade (shade distribution: A1-4; B1-4; C2-4; D2,4). The color appearance of a resin composite restoration is influenced and perceived by the translucency and light transmission characteristics (straight-line transmission and diffusion of light), as well as its colors (lightness, chroma, and hue)\(^{5,6,32,33}\). The background tooth color at the cavity floor can affect the color appearance of resin composite restoration, depending on the translucency of the restored composite. Additionally, at the border
of resin composite restorations, color-shifting can be caused on both the tooth and composite sides due to the color reflections of each other, which can be influenced by light transmission characteristics, especially the light diffusion transmission property of resin composites. Notably, ESQ has greater light diffusion transmission properties, while APX has lower light-diffusion but higher straight-line light transmission properties. Additionally, APX has higher transparency than ESQ (Table 2).

Regardless of their different light transmission properties and translucency, the color reproduction performance ($\Delta E_{00}$ between intact and restored teeth) was not significantly different between APX and ESQ. This might be due to the cavity size used in this study because, in the case of a shallow cavity, color-shifting is influenced by the straight-line, as well as diffused light transmission of the resin composite. However, $\Delta h^*$ was significantly different between APX and ESQ, although their minimum values were near-identical. This might be due to the different light transmission properties and/or translucency. Additionally, $\Delta L^*$ was significantly different between APX and ESQ, wherein both the minimum and maximum values in APX were greater than ESQ. These results indicate that APX is difficult to match in lightness to any tooth shade compared with ESQ. It is well-known that the most common shade matching failure between the resin composite and tooth is caused by a control error in lightness because the human eye is sensitive to light and dark, compared to color identification. In addition, it has been reported that nanofilled composites exhibit superior polishesability than conventional hybrid composites, leading to smaller surface roughness and higher initial gloss. Since the lightness of resin composites is inversely proportional to surface roughness and APX is a conventional microfilled hybrid composite, while ESQ is a nanofilled hybrid composite. This might be the reason why APX was inferior to ESQ in the $L^*$ color adaptation.

Nevertheless, the $\Delta E_{00}$ of OMN was significantly lower than those of APX and ESQ, as well as its $\Delta L^*$ and $\Delta h^*$ values. The higher performance of OMN in the color reproduction to various shades of teeth would be produced by the superior adjustment potential to the colors of the surrounding tooth substrates. OMN is a nanofilled hybrid composite that comprises 260 nm spherical fillers and has a higher straight-line light transmission property without light diffusion transmission. The manufacturer claims that the structural color phenomenon occurs due to the precisely uniform filler shape and size in OMN and that it could improve the color adjustment to surrounding tooth substrates.

Structural colors are produced by submicron-sized microstructures that reflect light at a specific wavelength through physical processes, such as optical interference. Since electronic excitation (absorption) of molecules is not involved in the mechanism of structural coloration, structural colors do not significantly fade unless the microstructure is destroyed. The jewel beetles, thin stratified layers with thicknesses of approximately 100 nm cause multilayer optical interference, which results in brilliant structural colors. Colloidal crystals, another example of structural colors that can be synthesized in a laboratory, consist of a periodic arrangement of small colloidal particles that appear brightly colored when the lattice period is comparable to the wavelength of light. Because the microstructure is the main factor affecting the wavelength of the reflection, structural colors are advantageous based on the premise that colors can be simply modified by adjusting the size of the microstructure without changing the overall material design, and many attempts have been made to apply colloidal crystals to produce different colors. However, these structural colors exhibit an angular dependence in the wavelength of the reflection. This is not desirable for general-purpose pigments, which are required to be the same color regardless of the viewing angle. These angular dependencies have been solved with hints from animals in nature: the feathers of some bird species possess less or non-iridescent structural colors. The microstructure of these features is neither periodic nor regular, but rather amorphous, which have a short-range order, thereby resulting in constructive optical interference at specific wavelengths. Furthermore, these structures are substantially isotropic. This implies that under diffuse illumination, reflections become almost angle-independent. Therefore, the amorphous arrangement of colloidal particles is essential for angle independent structural color materials. Recently, materials consisting of amorphous monodisperse silica particles which develop structural colors have been successfully prepared.

In the measurement of spectral reflectance, OMN generated a broad peak in the spectral reflection range of 430–750 nm (blue-to-red). On the other hand, a peak in the range of 430–450 nm (blue) was observed in the reflection spectrum R1 with fumed silica fillers of size 5–50 nm and R2 with 100 nm spherical fillers, and their reflectance gradually decreased in longer wavelengths. Generally, when light illuminates translucent materials, the shorter-wavelengths of the visible spectrum are reflected, whereas the longer-wavelengths are transmitted, giving the material a bluish appearance under reflected light. It is suspected that the 260 nm spherical fillers with a precisely uniform shape and size in OMN are responsible for the development of the structural color phenomenon in the yellow-to-red range by the effect of the amorphous aggregates.

In this study, the R1 and R2 composites were prepared at approximately the same contrast ratio and low light diffusion property of OMN to minimize the effect of light transmission properties and translucency on color adjustment. The $\Delta E_{00}$ of OMN in various-shade teeth was significantly lower than those of R1 and R2, and all the color parameters ($\Delta L^*$, $\Delta h^*$, and $\Delta C^*$ values) of OMN were significantly lower than those of the R1 and R2 composites. These results indicate that
the broad reflection spectrum through structural color phenomenon could improve the color reproduction performance of resin composite restoration in teeth of various shade. The structural color development in the yellow-to-red range by the light incident on the composite directly and/or through the surrounding enamel and the reflection color from dentin at the bottom of cavity might contribute to the improvement of color matching of the restoration in a complex manner.

On the other hand, there was also a significant difference in the \( \Delta E_0 \) between R1 and R2 composites in similar minimum values. These results implied that R1 had a poorer color adjustment potential in various-shade teeth than R2, even though the best color-matching performance to teeth were at a rather similar level between R1 and R2. Additionally, there were no significant differences in \( \Delta L^* \) and \( \Delta h^* \) between R1 and R2, but they differed considerably in \( \Delta C^* \). The R1 composite had a greater reflectance in the blue range than the R2 composite. These results might indicate that the greater reflection of the short wavelengths (blue range) could affect especially the \( \Delta C^* \) parameter of the composite restoration, leading to the poor color adjustment potential to the surrounding tooth tissues.

Based on the results of this study, the null hypothesis had to be rejected. The structural color phenomenon in the resin composite could improve the color adjustment potential to various-shade teeth. However, in OMN, the light diffusion and opacity are lowered to induce structural color phenomenon, which might generate another aspect of constructing an esthetic appearance of resin composite restorations. An increase in the light transmission property of composites can contribute to obscuring boundaries in resin composite restoration by the color-blending of resin composites and surrounding tooth substrates with each other\(^\text{12}\), and an increase in opacity of composites is necessary when masking the background tooth color is clinically required. Furthermore, the color adaptation of the resin composite restoration would be influenced by the size and depth of the cavity as well. Further studies are necessary to evaluate the color-blending effect of resin composites with structural color phenomenon at the border, as well as to investigate the color adaptation of the restored tooth with and without the need to mask the background tooth color, using various size and depth cavities.

CONCLUSION

Within the limitations of this study using human extracted teeth of various shades, it was concluded that OMN containing 260 nm spherical fillers developed the structural color phenomenon and a broad reflection spectrum, which could contribute to the improvement of the color adjustment of various-shade teeth. In contrast, the composites containing fillers of different shape or size did not seem to produce the structural color phenomenon, resulting in poor color adjustment of the restoration to various-shade teeth. Additionally, the greater reflection of the short wavelengths (blue range) would reduce the color compatibility of resin composite restorations.

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