A Comparative Evaluation of the Staining Capacity of Microhybrid and Nanohybrid Resin-based Composite to Indian Spices and Food Colorants: An *In vitro* Study

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Abstract

Introduction: Resin composite restorative materials can mimic the natural color and shade of the tooth. However, exogenous colorants from food and drinks can stain them due to adsorption. The influence of Indian food colorants and spices on resin composite restorations has not been evaluated extensively. **Aim:** This study aims to evaluate the staining capacity of microhybrid and nanohybrid resin-based composites, to saffron extract, tandoori powder, and turmeric powder. **Subjects and Methods:** Forty samples of microhybrid (Kulzer Charisma) and nanohybrid (3M Filtek Z350) resin composites were prepared using an acrylic template of dimension 5 mm × 3 mm. They were randomly divided into four groups and immersed into solutions of saffron extract, tandoori powder, and turmeric powder. Distilled water was used as the control group. Color values (L*, a*, b*) were measured by colorimeter using the CIE L*a*b* system before and after 72 h of immersion. Color differences ΔE*ab were statistically analyzed. **Statistical Analysis Used:** Two-way ANOVA and *post-hoc* Tukey (honest significant difference) test were done using IBM SPSS Statistics for Windows, Version 19.0. Armonk, NY: IBM Corp. **Results and Discussion**: All the immersion media changed the color of the resin composites to varying degrees. However, turmeric solution showed the maximum mean color variation ΔE*ab of 14.8 ± 2.57 in microhybrid resin composites and 16.8 ± 3.50 in nanohybrid resin composites. **Conclusion:** Microhybrid and nanohybrid resin composites tend to stain to Indian food colorants, especially to turmeric powder.

Keywords: *Colorimeter, color stability, microhybrid, nanohybrid, resin composite, spices, turmeric*

Introduction

Resin-based composite restorations require functional/biological as well as esthetic properties, such as good color matching and color stability.[1] A recent systematic review reported that anterior direct resin composites' restorations have a good longevity in long-term follow-up of 3+ years, with an annual failure rate of only 0%–4.1%. However, this review reports that one of the main reasons for failures of these restorations is unaesthetic appearance over a period. Color alterations, surface staining, and marginal discoloration are some of the esthetic failures.[2]

Many factors influence the color stability of the resin composites. Chemical interaction within the resin, water sorption, surface finish, and type of food colorants can cause color changes intrinsically and extrinsically.[3] Oxidation of the amine accelerator, oxidation in the structure of the polymer matrix, and unreacted monomers cause chemical discoloration which is intrinsic in nature. Hydrophilicity of the resin matrix contributes to the degree of water sorption into the resin matrix that leads to discoloration.[4] Exogenous colorants from food and beverages have been proven to discolor the resin composites by absorption and adsorption to the surface.[5] The compatibility of the colorant to resin matrix and the pH of the colorants are some influencing factors in this regard.[6]

Extrinsic surface staining is influenced by the surface finish and polishability which in turn is directly related to the type, content, and size of the filler particle.[7] Various modifications have been made in the filler particle size to obtain a good polishability and surface finish. Resin composites have been classified by Lutz and Phillips based on filler particle size, as macrofilled composites (particles from 0.1 to 100 μm), microfilled composites (0.04 μm particles),

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and hybrid composites (fillers of different sizes).[8] Hybrid composites combined the optical properties of the microfilled as well as the mechanical properties of the macrofilled composites. Furthermore, influence of nanotechnology in the dental materials has resulted in the development of nanocomposites. Nanohybrid composites are those that contain milled glass fillers and discrete nanoparticles (40–50 nm) as dispersed phase. They have been found to provide better mechanical strength and surface finish than hybrid composites.[9]

Numerous studies have been done on the effect of coffee, tea, red wine, and juices over different types of composite materials and differently processed composite restorations.[10-13] Although these beverages are common for all cultures, it is worthwhile to study the ethnic differences in food and its influence on esthetic restorative materials. Especially the Indian cuisine is abundant with flavors and colors, where natural and artificial colorants as well as spices are added. Relatively lesser studies are available in the literature assessing the effect of these colorants and spices on the resin composites.

Among these studies, more focus had been on turmeric powder. The results show that turmeric has a greater tendency to stain the resin composites.[14-17] Turmeric is fondly called in Indian cuisine as "kitchen queen," and because of its taste and color, it is called as "Indian Saffron" in Europe. Apart from color and taste, it is mostly added in abundance in Indian cuisine for its inherent therapeutic properties also. The therapeutic activity is attributed to the content of curcumin/curcuminoids, a polyphenol, which also contributes to the yellow color of the spice.[18]

Paprika and tamarind that are widely used in other Asian foods have also been evaluated recently. Saffron is one of the most expensive spices but a widely used one in Indian foods for flavor and color.[19] Saffron is the dried elongated stigmas and styles of the blue-purple saffron flower *Crocus sativus* L. The color, taste, aroma, and medicinal properties can be attributed to the bioactive compounds in saffron, mainly the carotenoids. The hydrophilic carotenoid in saffron called as Crocin is deep red. It readily dissolves in water to form an orange-colored solution; thus, it is widely used as a natural food colorant.[19]

Synthetic, intense orange-red color is yet a colorant added to the tandoori (clay-oven) foods of India. Tandoori foods are cooked under direct fire in clay ovens, called as "tandoor," used in Asian countries. The food cooked in this fashion usually uses strong red color to make it attractive. Natural food-coloring agents such as "annatto" seeds produce red-orange color to the tandoori foods. However, due to high cost and color stability, synthetic dyes have been favored over natural dyes globally. They are derived from petroleum or crude oil. As the health hazards of synthetic dyes are numerous, only six colors have been approved by the FDA. Red no: 40 (Allura red) and yellow no: 5 (tartrazine) and 6 (sunset yellow FCF) are used in commercial products to create food colors in this range.[20] No studies have been done so far on these colorants' effect on resin composites.

Thus, this study was conducted to assess the color stability of nanohybrid resin composites and hybrid resin composites to Indian food colorants and spices such as turmeric, saffron, and tandoori powder.

Subjects and Methods

Two resin composites of B2 shade used in this study are microhybrid resin composites (Charisma Smile, Universal light curing hybrid composite, Heraeus Kulzer, CE 0197, Lot-010105, Germany) and nanohybrid resin composites (Filtek Z350 XT, Universal restorative, 3M ESPE, CE 0123, Lot-1017 St. Paul, MN 55144-1000, USA). Table 1 shows the details of the materials used. Twenty cylindrical samples in each of the resin composites were prepared using an acrylic template in a dimension 5 mm × 3 mm. To obtain a flat and smooth surface and to avoid air bubbles, the material was placed in the template that was sandwiched between two glass plates lined with mylar matrices. It was then cured with LED light source (L1330232, Gulin WoodPecker, London, UK) with a wavelength 1000 mW/cm^2 for 20 s on each side. The tip of the curing unit was placed directly over the glass plate that was 2 mm thick glass slide; thus, the distance

Table 1: Materials used in the study

Composite resin	Technical specifications	Resin matrix	Filler loading (%)	Filler particle	Particle size
Nano hybrid	3M Filtek Z 350, USA B2 shade CE 0123 Lot-1017	Bis-GMA TEGDMA, UDMA, Bis-EMA	78.5	Zirconia Nano silica Cluster size	4-11 nm 20 nm 0.6-1.4 µm
Micro hybrid	Heraeus Kulzer Charisma, Germany B2 shade CE 0197 Lot-010105	Bis-GMA TEGDMA	78	Pyrogenic Silicone dioxide Microglass filler particles	0.01-0.07 µm 0.7-2 µm

Bis-GMA=Bisphenol A diglycidyl dimethacrylate, Bis-EMA=Bisphenol A ethoxylated dimethacrylate, TEGDMA=Triethylene glycol dimethacrylate, UDMA=Urethane dimethacrylate between the light source and samples was standardized. The output of the curing units was checked with a radiometer, L. E. D Radiometer (Kerr Manufacturing Products). The prepared samples were then wet ground using 1000 grit silicon carbide abrasive on one side manually for 10 s in running tap water along one direction.[21] They were polished in a planar direction using Shofu Super-Snap Mini Kit composite polishing kit (Finishing and Polishing composite restorations PN 0505, CE 0044 SHOFU INC., Kyoto 605-0983, Japan) sequentially from coarse discs to fine discs, on the one side only. The samples were stored in distilled water before subjecting to baseline color value assessment using colorimeter (CIE Lab observer-10° illuminant D65, Premier Colorscan SS 5100 A, CLRI, Chennai, Tamil Nadu, India). As a blinding process, the samples were randomized using "free online random generator tool."[22] The samples were positioned in the slot against white background in a colorimeter and the baseline l* a*b values were recorded. Colorimeter measures the amount of light reflected by the selected colors and the color measurements are based on the CIE lab system, developed by Commission Internationale d'Eclairage for characterizing colors based on human perception. A standard illuminant A against a white background was used in this.[23]

A 0.1% solution of turmeric, saffron, and tandoori powder was made with distilled water. The resin samples were immersed for 72 h in the test solutions and distilled water, which was used as the control (Group W – Water immersion [n = 5]; Group S – Saffron immersion [n = 5]; Group T – Tandoori color powder [n = 5]; Group TP – Turmeric powder [n = 5]). The solutions were changed every day and were agitated periodically. After 72 h of immersion, the samples were again tested in colorimeter to evaluate the changed color values. An average of three repeated readings was obtained from each sample.

The color difference (ΔE*ab) before and after immersion was calculated using the formula $\Delta E^*ab = ([\Delta L]^2 + [a]^2 + [b]^2)^{1/2}$. Two-way ANOVA and *post hoc* Tukey (honest significant difference [HSD]) test were done using IBM SPSS Statistics for Windows, Version 19.0. Armonk, NY: IBM Corp, USA.

Results

Table 2 shows the mean and standard deviation in the color difference before and 72 h after immersion in the test solutions. The mean Δ E*ab value obtained with turmeric solution for microhybrid composite and nanohybrid composite resin was the highest among all the solutions. It was 16.8 ± 3.50 and 14.7 ± 2.57, respectively. Resin samples immersed in saffron and orange-red synthetic color used in tandoori foods also showed color changes as that of distilled water group. Table 3 shows the two tailed t test for intergroup comparison. There was no significant difference found with the immersion medium. Table 4 and 5 shows the two-way ANOVA and Post-hoc Tukey test results. No significant difference was observed on the interaction between the resin types and the solutions at $P = 0.208$. Neither there was a significant difference between the composites in its capacity to be stained at $P = 0.739$. However, a significant difference was observed in the capacity of the solutions to stain both the composites at $P < 0.01$. This multiple comparison elicited that the color difference, ΔE*ab values observed in samples immersed in turmeric solution were statistically highly significant when compared to all the other solutions in both hybrid and nanohybrid resin samples. The ΔE*ab values for samples immersed in saffron and tandoori color solutions were not statistically different from the distilled water group or with each other.

Table 2: Mean and standard deviation of Delta E

Immersion media	Composite resin	Delta E (mean±SD)
Distilled water	Hybrid	1.2751±0.40220
	Nanohybrid	0.8132±0.73868
Saffron solution	Hybrid	0.8302±0.54090
	Nanohybrid	1.2044±0.45141
Tandoori color solution	Hybrid	1.0804±1.15572
	Nanohybrid	2.4440±2.11175
Turmeric solution	Hybrid	16.8056±3.50206
	Nanohybrid	14.7652±2.57147

SD=Standard deviation

Table 3: Two-tailed *t*-test for intergroup comparison between the composite resins for each immersion medium

Immersion media (in hybrid vs. nanohybrid composite resin)	df	*t*	Two-tailed *P*
Distilled water	8	1.228	0.2544 (NS)
Saffron solution	8	1.188	0.2690 (NS)
Tandoori color solution	8	1.267	0.2409 (NS)
Turmeric solution	8	1.050	0.3244 (NS)

NS=Not significant

Discussion

The current study evaluated the ability of Indian spices and food colorants to stain the microhybrid and nanohybrid resin composites. The results of the study show that turmeric stained the resin composites more intensely than saffron and tandoori color powder. The color difference (ΔE) produced in the resin samples after immersion in turmeric solution for 72 h was much higher than the clinically acceptable level. The difference was also statistically significant compared to the other study solutions. This is in accordance with similar studies done with indigenous spices where turmeric was found to stain the resin-based composites more than tamarind, paprika, and tobacco.[16,17]

Table 4: Two-way ANOVA results for color change ΔE

	Sum of squares	Mean squares	*F*	*P*
Composite resins	0.365	0.365	0.113	0.739 (NS)
Immersion medium	1582.807	527.602	163.118	<0.001 (S)
Composite resin × immersion medium	15.574	5.191	1.605	0.208 (NS)

NS=Not significant, S=Significant

Table 5: *Post hoc* Tukey (honest significant difference)

Multiple comparison	Mean difference	SE	Significant
Distilled water			
Saffron solution	0.0268	0.80430	1.000
Tandoori color solution	−0.7180	0.80430	0.809
Turmeric solution	−14.7412*	0.80430	0.000
Saffron solution			
Distilled water	−0.0268	0.80430	1.000
Tandoori color solution	−0.7449	0.80430	0.791
Turmeric solution	−14.7681*	0.80430	0.000
Tandoori color solution			
Distilled water	0.7180	0.80430	0.809
Saffron solution	0.7449	0.80430	0.791
Turmeric	−14.0232*	0.80430	0.000
Turmeric solution			
Distilled water	14.7412*	0.80430	0.000
Saffron solution	14.7681*	0.80430	0.000
Tandoori color solution	14.0232*	0.80430	0.000

Based on observed means. The error term is mean square (error)=3.234. *The mean difference is significant at the 0.05 level. SE=Standard error

In the current study, saffron also caused discoloration of both composites after 72 h of immersion, if not to the clinically unacceptable level, at least to clinically perceptible level. In nanohybrid samples, there was a color difference of ΔE as 1.20 after immersion for 72 h in solution of saffron. So far, no study has been done on this aspect of saffron on resin composites color stability.

A synthetic orange-red food color preparation that is commonly used for tandoori foods was tested. The results of the study show that this colored solution stained both the resin types more than the saffron solution. There are no similar studies yet with regard to this.

The reasons for the change in color of the composites tested in this study to various spices can be attributed to many variables: (1) characteristics of the colorant, (2) type of matrix in the resin, and (3) filler content and type in the resin.

Polymers such as polymethyl methacrylate are usually colored using dyes and pigments. Organic pigments such as azo pigments that are in the yellow, orange, and red color ranges are highly compatible with polymer.[24] Curcumin obtained from turmeric and carotenoids obtained from saffron are organic pigments. They are polyphenols, nonpolar in nature; thus, they are hydrophobic.[16] Studies have shown that colorant solutions such as coffee and tea that have high polarity first leach out and discolor the resin matrix. They stain the surface of the resin or penetrate the resin by means of absorption and adsorption.[25] However, as curcumin and carotenoids are polyphenols that are not polar, polarity of the substance might not be the mechanism behind discoloration with these compounds. Instead, it is possible that these colorants disperse into the resin matrix as separate phase and thus discolor the resin.[24] Among the two, carotenoids are observed to color the resin monomer to a lesser extent compared to curcumin. The reason for deep intense coloring of curcumin-containing turmeric is not well understood and is worthy of further exploration.

The presence of triethylene glycol dimethacrylate (TEGDMA) in the resin matrix has been attributed to be one of the causes for discoloration of the resin matrix. TEGDMA is a diluent monomer added to the bisphenol A diglycidyl dimethacrylate resin matrix. Due to its hydrophilicity, composites containing this monomer exhibit increased water sorption.[9] Both Filtek Z 350 (nanohybrid composites) and Kulzer Charisma (microhybrid composites) tested in this study contain TEGDMA in their composition, which can be attributed to their similar color stability to various aqueous immersion media.

Resin composites with small filler particles are assumed to provide best surface finish and gloss thus have more color stability. Therefore, nanocomposites with nanoparticle as fillers are expected to show better color stability. However, in this study, both the microhybrid and nanohybrid groups show similar color stability. This can be explained by the type and size of filler particles in both composites. Filtek Z 350, the nanohybrid in this study, contains a combination of individual 20 nm silica nanoparticles and zirconia/silica nanoclusters of particle size 0.6–1.4 µm. The Heraeus Kulzer Charisma the microhybrid tested is composed of X-ray opaque microglass of size 0.7 µm and highly dispersive silicon dioxide of size 0.01–0.07 µm. It is interesting to note that the nanocluster size in the nanohybrid and the microglass size in the microhybrid are similar, 0.6 and 0.7 µm, respectively. Thus, both particles are prone for dislodgment leaving the surface equally rough and susceptible to staining.[16]

This experimental study has not taken into consideration the color changes assessment at periodic interval. This assessment would have given an insight into the rate of discoloration of the resin composites to the stains. Studies have reported composites and stains to behave differently with time of immersion.[24] The real consumption, amount, and frequency of these spices may vary with food habits and culture. Especially the inclusion of saffron in cuisine is less frequent when compared to turmeric and tandoori color powder. Thus, standardization to 0.1% was done based on previous study.[16] For the same reasons, the results of this study may not be valid enough to apply to clinical situations. Further clinical evaluation is essential. In addition, immersions in static solutions do not represent the dynamic nature of the oral environment. Thus, aging process should be included in further studies along with standardizing the temperature and agitation. The pH of the solutions, which might have effect on the surface degradation of the composites, and polishing procedures, which might have effect on the stainability, were not included in the current study. With no available evidence on saffron and synthetic colors used in Indian cuisine, study of the above-mentioned variables is imperative to validate the evidence generated by the current investigation.

Conclusion

Within the limitations of the study, it can be concluded that Indian spices such as turmeric and saffron and tandoori powder have the capacity to stain the resin composites. There was no significant difference in the staining capacity of the microhybrid and nanohybrid resin composites to these colorants, despite the difference in the filler technology. Further study is required to correlate the other variables such as time, temperature, aging and pH, concentration of the staining solutions. Characterization of the staining elements in these spices and understanding the mechanism of discoloration are mandatory to ensure long-lasting esthetic restorations in the Indian context.

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Conflicts of interest

There are no conflicts of interest.

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