Effect of Green Tea (*Camellia sinensis*) Solution on Color Change of Silorane- and Methacrylate-Based Composite Resins

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Effect of Green Tea (Camellia sinensis) Solution on Color Change of Silorane- and Methacrylate-Based Composite Resins

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Abstract. Color change is an undesirable but avoidable effect in composite resin. Limited research has been conducted on the effect of green tea solution on silorane- and methacrylate-based composite resins. In this study, we used 30 silorane- and methacrylate-based composite resin disks (shade A3) (6 mm in diameter and 2 mm in thickness) prepared from Filtek P90 and Filtek Z250. Specimens were divided into six groups (n = 5), and each group was immersed in three green tea solutions at different concentrations (1%, 2%, and 3%) for 6 hours. Color assessment was performed with a spectrophotometer based on CIE*Lab theory, and results were subjected to statistical analysis using one-way ANOVA and independent t-test. Significant differences were observed in ∆* between the two composite disks after immersion in tea solutions (p < 0.05). Color change in silorane-based composite resin was higher than that in methacrylate-based resin; this color change was the highest when silorane-based composite resin was immersed in 3% green tea solution (∆E = 3.85).

1. Introduction
Methacrylate-based composite resin has been the most common type of composite resin in the market. However, it has many limitations [1], of which the most prominent is the high value of polymerization shrinkage, which results in various complications, such as leakage, secondary caries formation, and pulp inflammation [2].

To fix these shortcomings, silorane-based composite resin was introduced [3]. One of the products that have been circulating in the market is Filtek P90 (3M ESPE). Silorane-based composite resin has a lower polymerization shrinkage value than methacrylate-based composite resin. The siloxane and oxirane molecules present in this composite resin form a ring-opening polymerization reaction [4], which reduces polymerization shrinkage.

Color change has an undesirable effect on composite resins; however, composite resin has a tendency to change color in the oral cavity [5,6]. Color change is the main reason why patients opt to change the restoration, as it is no longer has an aesthetic appeal. Color changes in composite resins are caused by intrinsic as well as extrinsic factors [7]. Intrinsic factors include chemical changes occurring within the resin matrix or changes in the matrix-filler bond [8], whereas extrinsic factors include the adhesion of stains on the composite resin surface or the absorption of colored liquids consumed on a daily basis [9]. Research shows that composite resins change color when exposed to a variety of dyeing agents, especially red wine, coffee, soft drinks, and tea [10, 11].

Today, green tea is one of the most widely used herbal ingredients. It is a very popular beverage that is consumed daily in Asia [12]. Its high popularity is due to many health benefits, including
antioxidant, anticaries, antibacterial, antiviral, antidiabetic, antimutagenic, and antitumoral effects [13]. However, regular tea consumption can discolor the restoration.

Although the influence of green tea solution on teeth and composite resins has been studied, limited research has been conducted on the effect of green tea solution on the color of silorane- and methacrylate-based composite resins. In this study, we investigated the color changes in both types of composite resins due to green tea.

2. Methods

This study was conducted at the Dental Materials Laboratory, Faculty of Dentistry, Universitas Indonesia from August to October 2014. Specimens consisted of 30 composite resin cylinders, including 15 silorane-based composite resins (Filtek P90, 3M ESPE, USA) and 15 methacrylate-based composite resins (Filtek Z250, 3M ESPE, USA). All specimens were sliced into cylindrical metal moulds (6 mm diameter and 2 mm height), which were coated with Mylar Strips and preparatory glass above which a load of 1 kg was placed. Lighting was done for 40 s using Light Curing Unit (Hilux, Benioglu, Europe) at a distance of ±1 mm. Subsequently, all specimens were immersed in aquadest solution for 24 hours for perfect polymerization, and the initial color was measured using the Vita Easy Shade intraoral spectrophotometer, with white paper as the base.

Each specimen was inserted into different plastic pots, and each pot was filled with 1%, 2%, or 3% green tea solution prepared by diluting 100% green tea extract with distilled water. Specimens were randomly assigned pots containing 5 ml green tea solution at different concentrations, with one specimen per pot (n = 5 for each concentration). Each pot was labeled based on the green tea concentration and the type of composite resin. Specimens were submerged in green tea solution for 6 hours at 37°C. Subsequently, specimens were removed and rinsed with aquades for 15 s, dried, and evaluated for color change. Differences in color change of composite resin were analyzed using one-way ANOVA and independent t-test at a significance level (α) of 0.05.

3. Results

In this study, silorane- and methacrylate-based composite resins were immersed in green tea solutions at different concentrations (1%, 2%, and 3%) for 6 hours at 37°C, and the change in resin color was measured using Vita Easy Shade. Using this tool, values of L*, a*, and b* were determined before and after immersion in green tea solution, and values of ΔL*, Δa*, and Δb* were calculated (Table 1).

| Composite resin | Concentration of green tea solution | ΔL* | Δa* | Δb* | ΔE* |
|-----------------|------------------------------------|-----|-----|-----|-----|
| Methacrylate    | 1%                                 | -0.64 | -0.02 | -1.80 | 1.94 |
|                 | 2%                                 | -2.70 | -0.18 | -1.12 | 2.82 |
|                 | 3%                                 | -0.52 | -0.40 | -1.12 | 1.37 |
| Silorane        | 1%                                 | -2.88 | -0.08 | 1.08  | 3.31 |
|                 | 2%                                 | -1.22 | -0.54 | 0.76  | 1.71 |
|                 | 3%                                 | -3.56 | -0.20 | 1.42  | 3.85 |

According to the concept of CIE, ΔE*, the difference between two colors was calculated using the formula ΔEab* = [(ΔL*)2 + (Δa*)2 + (Δb*)2] ½. Values of ΔE* ≤ 3.3 are clinically acceptable. Figure 1 shows values of ΔE* of silorane- and methacrylate-based composite resins soaked in green tea solution for 6 hours. Results showed that in the case of methacrylate-based composite resin, the highest ΔE* value was observed in 2% tea solution, followed by 1% and 3%...
solutions; in the case of silorane-based composite resin, the highest ΔE* value was obtained in 3% tea solution and the lowest in 2% solution.

**Figure 1.** Graph showing average color changes (ΔE*) of silorane- and methacrylate-based composite resins after immersion in green tea solution.

Statistical analysis using one-way ANOVA revealed that ΔE* values were significant for all three concentration groups ($p = 0.010$) (Table 2).

| p-values | Information  |
|----------|--------------|
| Green Tea 1% | Green Tea 2% | 0.044* | Significant |
| Green Tea 3% | 0.168 | Not significant |
| Green Tea 1% | Green Tea 1% | 0.044* | Significant |
| Green Tea 2% | Green Tea 3% | 0.003* | Significant |
| Green Tea 1% | 0.168 | Not significant |
| Green Tea 2% | Green Tea 2% | 0.003* | Significant |

* $p < 0.05$

One-way ANOVA tests were also performed on silorane-based composite resins immersed in 1%, 2%, and 3% green tea solutions to determine the difference in E* (ΔE*) values. Results revealed statistically significant differences at all three concentrations of green tea ($p = 0.002$). The highest color change value was observed in the 3% green tea (Table 3), and no significant differences were observed in ΔE* between 1% and 3% green tea.

**Table 3.** Statistical significance of color change (ΔE*) of silorane-based composite resins in different concentrations of green tea solution.

| p Value | Information  |
|---------|--------------|
| Green Tea 1% | Green Tea 2% | 0.005* | Significant |
| Green Tea 3% | 0.262 | Not Significant |
| Green Tea 1% | Green Tea 1% | 0.005* | Significant |
| Green Tea 3% | 0.001* | Significant |
| Green Tea 1% | 0.262 | Not Significant |
| Green Tea 2% | Green Tea 2% | 0.001* | Significant |

* $p < 0.05$
Differences in $\Delta E^*$ values of silorane- and methacrylate-based composite resins were analyzed using an unpaired $t$-test. Results showed that differences in $\Delta E^*$ values between silorane- and methacrylate-based composite resins were statistically significant ($p = 0.020$).

4. Discussion
Results of this study showed significant differences in $\Delta E^*$ values between composite resins immersed in 1%, 2%, and 3% green tea solutions. An increase in the value of $\Delta E^*$ of methacrylate-based composite resin was observed from a concentration of 1% to 2% tea solution; however, $\Delta E^*$ value decreased at 3% concentration. In the case of silorane-based composite resin, the least change in color was observed in 2% tea solution and the greatest change in 3% concentration. These differences in color change could be influenced by various intrinsic and extrinsic factors.

The intrinsic factors affecting these results could be water absorption by matrix as well as oxidation in the amine accelerator [7]. An extrinsic factor that could potentially affect our results is the tannin content of green tea solution; tannin is a polyphenol compound that is yellow to brownish in color [14].

Composite resin has a tendency to absorb water and other colored liquids, which causes discoloration [7, 15]. Silorane-based composite resins are new composite resins that utilize organic matrices of siloxane and oxirane. It has been shown to be more resistant to color change, as it has chemically stable when immersed in liquid medium, has a low water absorption rate, low solubility, and low diffusion coefficient [16].

However, in this study, the highest color change was observed in silorane-based composite resins soaked in 3% green tea solution with $\Delta E^* = 3.85$, which exceeded the clinically acceptable range [17]. Statistical analysis showed that this value was significant, which is consistent with results of Barutcigil et al. showing that the discoloration of silorane-based composite resins is higher than that of methacrylate-based composite resins after immersion in aquadest for 1 month [7].

We showed that $\Delta E^*$ values of methacrylate-based composite resins differed significantly between 1% and 2% and between 2% and 3% tea solutions. In methacrylate-based composite resin, the largest color change occurred in 2% tea solution. However, all the color changes that occurred in methacrylate-based composite resins were still clinically acceptable. In silorane-based composite resins, there was a significant difference between $\Delta E^* 1\%$ and $2\%$ and $2\%$ and $3\%$ with $\Delta E^* 1\%$ and $3\%$ unacceptable clinical concentrations. In contrast, the $2\%$ concentration group has the lowest $\Delta E^*$ value. This was probably because the color change is heavily dependent on the degree of polymerization that occurs in the composite resin [3].

Color changes in silorane-based composite resins were higher than those shown previously; silorane-based composite resin is expected to undergo a lower color change because of its hydrophobic matrix [3]. The difference in matrix structure possibly affected the polymerization reactions to occur differently. In silorane-based composite resins, polymerization reactions are called ring-opening reactions; in these reactions, each monomer approaches the nearest monomer, and the monomer ring is then exposed to form a polymer. This process takes longer than polymerization reactions in methacrylate-based composite resins. The number of unreacted monomers makes the polymer matrix heterogeneous [18]. This phenomenon decreases the density of crosslinks in the matrix and increases the water absorption capacity of the resin [18].

Water absorption by the matrix also causes hydrolytic degradation and loosens the bonds between filler particles and matrix resin. This hydrolytic degradation process starts from the ions contained in the filler. Some ions have electropositive properties and tend to react with water, which disturbs the stability of silica bonds and the space created by ions is replaced by hydrogen ions from water. Increased hydroxyl ion concentrations cause siloxane bonds (Si-O-Si) to break down, thus causing surface degradation [19]. Overall, we conclude that the color change in silorane-based composite resin was due to the high rate of water absorption and the breakdown of covalent bonds between the filler and matrix. If the polymerization occurs perfectly, hydrophobic properties of the silorane matrix improve color stability [3].
This result was in line with that of Beurgers et al. in Heshmat showing that in immersion in the aquades for seven days, the stability of the color of the silorane-based composite resin was low [3]. Based on micrographic assessment, Pires-de-souza et al. have shown that quartz particles in silorane-based composite resins (Filtek P90) detach from the resin matrix during immersion [4].

The methacrylate-based composite resins used in this study contained Bis-GMA, which is known to have a 3%-6% water absorption rate [17]. Bis-GMA is more susceptible to discoloration than UDMA. Water absorption by methacrylate-based composite resin was evident from its reduced brightness due to the tannin content of green tea solution. The green tea extract used in this study contained 15.21% tannin. Tannin present in green tea is condensed and water-soluble [20]; the water-soluble tannin causes the color of the resin to darken. Upon water absorption, the bond between the matrix and filler in the composite resin is damaged, generating microcracks [7] through which tannin penetrates the resin and causes discoloration [21].

The filler used in the resin can also affect its discoloration [7]. The detachment of filler from matrix bonds increases the empty space in the composite surface, which increases surface roughness and susceptibility to extrinsic stains [22]. Additionally, the pH of green tea solution used can have an effect. Composite resins immersed in a solution with a low pH exhibit a coarser surface than those soaked in a solution with high pH, as low pH could speed up the hydrolysis reaction [23].

An intrinsic factor that affects the color change of resin is the presence of the photoinitiator, camphorquinone (CQ). It is the most common photoinitiator used in composite resins and serves to accelerate the polymerization process. Luiz BKM et al. in Barutcigil and Yildiz have shown that CQ has a significant effect on the resin, even when used in small amounts [7]. If CQ is not fully polymerized because of inadequate irradiation, it imparts a yellowish color on the composite resin [11]. Additional photoinitiators, such as tertiary aromatic or aliphatic amines, accelerate resin discoloration. Amines of all kinds induce yellow or brownish discoloration under the influence of light and heat (Widow et al. 2004). This discoloration is permanent and reduces the aesthetics of composite resins [11].

5. Conclusion
This study showed differences between silorane- and methacrylate-based composite resins in color change when immersed in 1%, 2%, and 3% green tea solutions. Color change was higher in silorane-based composite resin than in methacrylate-based composite resin.

6. References
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