Effect of nanofilled self-adhesive protective coating on color changes and surface roughness of composite resin

Margareta Rinastiti, Andina Widyastuti**

** Department of Conservative Dentistry, Faculty of Dentistry, Universitas Gadjah Mada, Yogyakarta, Indonesia*

Correspondence: rinastiti@ugm.ac.id

Received 13 October 2021; 1st revision 11 May 2023; 2nd revision 5 July 2023; Accepted 11 July 2023; Published online 31 July 2023

Keywords:
Aging; composite-resin; discoloration; nanofilled self-adhesive protecting coating; surface roughness

ABSTRACT

Background: *Discoloration of composite restorations may affecting aesthetic appearance. The aging-process and surface roughness may influence color changes. A nanofilled self-adhesive protecting coating has been developed for coating tooth restoration and expected to prolong the longevity of restoration. To evaluate the effect of nanofilled self-adhesive protective coating on the surface roughness and color-changes of flowable and packable composite after aging condition.*

Method:*The total of 40 discs (15x2mm) and 60 boxes (20×10× 2mm) specimens from flowable and packable composite were used. A half of the specimens was coated by using protective-coating. The reflectance chromameter was used to measure the color baseline. Afterward, the specimens were subjected into aging process by immersing in artificial saliva and carbonated drink (37°C,7d). The color changes were calculated based on the [CIE L*a*b*].For surface roughness, box-shaped specimens were measured in fresh condition, after aging conditions.*

Results:*The color change ranged from 0.49 to 2.31. Applying protective-coating was associated with a significant decrease in the color-changes and surface roughness after aging. The changes of three color coordinates resulted in significant differences for both composite, with and without protective coating application.*

Conclusion:*The application of protective-coating decrease the color-changes and surface roughness of flowable and packable composite-resin.*

doi: http://dx.doi.org/10.30659/odj.10.1.28-36
2460-4119 / 2354-5992 ©2022 National Research and Innovation Agency

Odonto : Dental Journal accredited as Sinta 2 Journal (https://sinta.kemdikbud.go.id/journals/profile/3200)
How to Cite: Rinastiti *et al*. Effect of nanofilled self-adhesive protective coating on color changes and surface roughness of composite resin. Odonto: Dental Journal, v.10, n.1, p. 28-36, July 2023.

INTRODUCTION

Composite-resin (hereafter: composite) is the most widely used dental material due to its superior physico-mechanical and aesthetic properties. However, because to the dynamic oral environment, composite may suffer and need to be repaired or replaced after being exposed to the oral cavity.[1] Esthetic failure of tooth restoration that is caused by discoloration is one of the major reason to replace the restoration.[1, 2] As a result, the color stability of composite materials is critical, whether after curing or over the life of the restoration.

Discoloration of composite restoration were influenced by intrinsic and/or extrinsic factors. Absorption of stains comes from food and beverage, incomplete polymerization, chemical reactivity, microorganism, oral hygiene, and restoration's surface topography are categorized as extrinsic factors.[3–5] While, intrinsic factors involve the interaction of filler, matrix or silane coupling agent. When the materials are aged under the dynamic oral situation, such as thermal changes, humidity, and ultraviolet exposure, the discoloration will be penetrated in the deeper portion.[6]

The study of the aging process of the composite-resin is conducted by imitating a state in the cavity mouth through a number of in vitro assays, namely storage in saliva,[7-9] immersion in citric acid, carbonated drink and thermocycling.[10,11] or prone by using biofilm.[12] The aging process can lead to the degradation of composite and may affect the color stability. Water storage and immersion in citric acid resulted in hydrolysis and elution of filler particle and water resorbtion into matrix.[11,13] Carbonated beverages are a type of soft drink that typically contain additional acidity regulators, such as malic acid, citric acid, or phosphoric acid, as well as sweeteners and sugars. These drinks are known to have a significant impact on the risk of dental caries and the degradation of composite restoration compounds.[14] The degradation of composite restorations can be influenced by carbonated soft drinks through the mechanisms of acid attack and water sorption. The higher solubility of composite resin in acidic solutions may lead to surface erosion, which could potentially increase surface roughness and diminish the aesthetic quality of the restoration.[15] Matrix dissolution and the interface failure between matrix and filler particle may result in composite discoloration.[16] It was evaluated that chemical dissociation of the resin matrix itself and/or matrix-filler interface over time are reported to be the main causes of intrinsic discoloration.[17] In addition, the in vitro aging method enhanced the composite's roughness.[18,19] The surface's irregularity may manifest discoloration as a result of the absorption of ingredients from beverages and food·.[17]

A material-based matrix and nanofiller have been developed for coating glass ionomer restorative materials cement and composite- resin. This liquid offers potential advantage such as simple application, additional protection for restoration-tooth margin, improve wear resistance, and inhibit the deposition of stain and plaque formation. Moreover, a polished restoration may be achieved in a less clinical steps.[20] Previous studies have confirmed that the application of uniformly dispersed film thickness (35 to 40 μm) on the restoration surface provides higher wear resistance, strengthen restoration and reduce microleakage.[21] Very few studies have examined the effect of nanofilled self-adhesive protecting coating on the composite restoration performance.

The purpose of this research was to evaluate the effects of nanofilled self-adhesive protective coating on the surface roughness (Ra) and color changes (ΔE) of flowable and packable composite-resin after aging condition. The null hypothesis tested was that the color changes and surface roughness of the aged flowable and packable composites that was coated with the nanofilled self-adhesive protective agent were lower than those of composites without a protective coating.

RESEARCH METHOD

This study was approved by the Ethics Committee, Faculty of Dentistry, Universitas Gadjah Mada No. 00795/KKEP/FKG-UGM/EC/2016. The flowable and packable microhybrid composites were used in this study is presented in Table 1.

Table 1. Materials used in the research

Material	Composition	Manufacture
Filtek supreme flow Z350XT™(low viscosity, nanoparticle-filled composite	Matrix : bis-GMA, TEGDMA, and Procrylat K Filler : ytterbium trifluoride filler with a range of particle sizes from 0.1 to 5.0 µm, a non-agglomerated/non-aggregated surface-modified 20 nm silica filler, a non-agglomerated/ non-aggregated surfacemodified75nmsilicafiller, and surface-modified aggregated zirconia/silica cluster filler (comprised of 20 nm silica and 4 to 11 nm zirconia particles). The aggregate has an average cluster particle size of 0.6 to 10 microns (65 % wt, 47 % vol)	3M ESPE
Filtek supreme Z350 XT™ (nanoparticle-filled composite)	Matrix : Bis-GMA, Bis-EMA, UDMA, Bis-PMA, DUDMA, TEGDMA Filler : non-agglomerated/non-aggregated 20 nm silica filler, non-agglomerated/non-aggregated 4 to 11 nm zirconia filler, and aggregated zirconia/silica cluster filler (comprised of 20 nm silica and 4 to 11 nm zirconia particles), average cluster particle size of 0.6 to 10 microns (82 % wt, 60 % vol)	3M ESPE
G Coat™	Multifunctional urethane methacrylate, metil metacrylate, Aliphatic dimetacrylate, Tertier amine, champorquinone, silicon dioxide, phosphoric ester monomer	GC

Specimen Preparation

Two hundred specimens (100 specimens of each restorative material) were prepared by filling a plastic ring mold (10 mm in diameter and 2 mm in thickness, for color measurement) or box mold (20 × 10 × 2) mm, for surface roughness measurement). Both sides of uncured composite-resin were covered with celluloid strip and pressed flat with a microscopic glass slide. All specimens were polymerized for 20 s with an LED curing light. For each composite-resin type, 20 specimens were treated with and 20 specimens without nanofilled self-adhesive protective coating. A half of the specimens from each composite were then aged by immersing in artificial saliva, while the remaining specimens in the carbonated drink for 7 d at 37 °C. The evaluation of color changes and surface roughness were evaluated prior to- and after aging conditions.

Evaluation of Color Changes

The chromameter were used to measure the color of all specimens, based on the *Commission Internationale de l'Eclairage* [CIE L*a*b*] system.[22]

The colors were measure at the baselines (T0) and at a time interval of 7 days (T7) for the corresponding material. A tissue paper was used to wipe the specimens. The dry specimens were placed in the viewing port of the chromameter. L*, a* and b* values, where "L" namely white-black, "a" red-green, and "b" yellow-blue. The spectrophotometer was automatically calculated and recorded the mean values of Δ L*, Δ a*, Δ b* after three measurements. The color difference (ΔE) was calculated from the mean ΔL*, Δa*, Δb* values for each specimen using the following Formula (1) :

$$\Delta E^*_{ab}\,(L^*a^*b^*) = [(\Delta L^*)^2 + (\Delta\ a^*_{ab})^2 + (\Delta\ b^*_{ab})^2]^{½} \quad (1)$$

Evaluation of Surface Roughness

The roughness of composite surface readout was made over a distance of 5 mm with a cut-off of 0.8 mm, at a speed of 0.25 mm/s. Three measurements were taken at different sites on the specimen surface to calculate the mean of composite roughness (Ra).

RESULTS

Color Changes

The means and standard deviations for values of ΔE^*_{ab} of flowable and packable composite with and without nanofilled self-adhesive protective coating are shown in Table 2 and Figure 1.

Table 2. The mean (ΔE) and standard deviation of composite resin color changes prior to- and after saliva and carbonated drink immersion

	FS	FC	PS	PC
Without nanofilled self-adhesive protective coating	2.15 ± 0.47^{a}	2.31 ± 0.66^{a}	1.04 ± 0.33^{b,d}	1.48 ± 0.74^{b}
With nanofilled self-adhesive protective coating	1.24 ± 0.69^{b,d}	1.34 ± 0.65^{b}	0.49 ± 0.22^{c}	0.80 ± 0.29^{c,d}

FS : Flowable composite resin, after saliva immersion
FC : Flowable composite resin, after carbonated drink immersion
PS : Packable composite resin, after saliva immersion
PC : Packable composite resin, after carbonated drink immersion
Different letters are showing significant differences

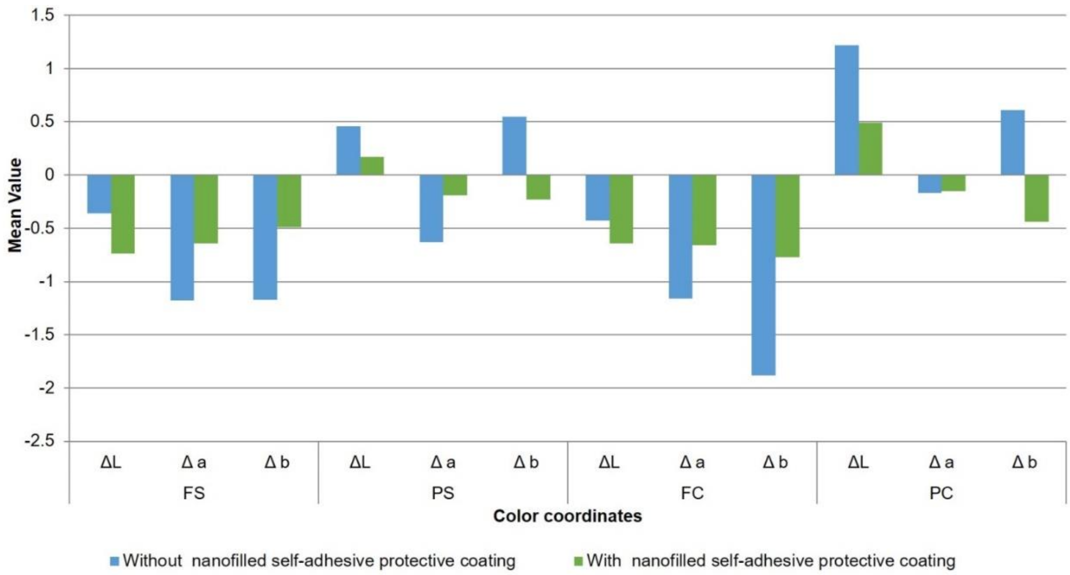

Figure 1. Mean values of color coordinates of different composites and surface treatment after saliva and carbonated drink immersion.

Table 2 showed that the mean color changes of both flowable and packable composites without nanofilled self-adhesive protective coating were significantly larger ($p < 0.05$) compare to the composites that were protected by nanofilled self-adhesive protective coating agent. The color changes of the flowable composite were smaller than packable composite with or without nanofilled self-adhesive protective. There were insignificant differences between the aging method for all composites ($p > 0.05$, Table 2).

After a period of 7d immersion, the ΔL^* for all the composites, surface treatments and aging methods ($p < 0.05$) were significantly change. The lighter specimens were indicated by positive ΔL, whereas negative ΔL^* indicates that the specimens became darker. The flowable composite was darker than packable composite for all aging method (Figure 1).

A significant change of Δa^* was revealed by all composite-resin after 7 d aging either in the saliva or carbonated drink ($p < 0.05$) for all of the groups. Negative Δa^* shows a shift towards green color. A significant change also noted for the changes along yellow-blue axis (Δb). Flowable composite showed the color changes toward blue, while the positive value of packable composite-resin indicated that the composites were shifted toward yellow color either after saliva or carbonated drink immersion.

The correlation between color changes and brightness showed moderate negative linear correlation (-0.31) There were strong correlations between color changes along red-green axis (-0.62) and along yellow-blue axis (0.58). The significance value of Brightness (ΔL), change along red-green axis (Δa), change along yellow-blue axis (Δb) are $p = 0.006$, $p = 0.000$ and $p = 0.000$ respectively.

Surface Roughness

Table 3 presents the surface roughness (Ra) of composite-resin. There was a significant different among the composite, surface treatments and aging method ($p < 0.05$). It was verified that the composites that were not coated with nanofilled self-adhesive protective coating presented higher surface roughness in comparison with composite with coating agent. With regard to composite with coating agent, there was no significant difference for both aging methods, except for packable composite after saliva immersion ($p > 0.05$).

Table 3. The mean of surface roughness (Ra) and standard deviation of composite resin color changes prior to- and after saliva and carbonated drink immersion

	F	FS	FC	P	PS	PC
Without nanofilled self-adhesive protective coating	0.03 ± 0.01	0.43 ± 0.01	0.14 ± 0.06	0.54 ± 0.23	1.77 ± 0.66	1.77 ± 0.6
With nanofilled self-adhesive protective coating	0.03 ± 0.01	0.04 ± 0.01	0.04 ± 0.01	0.40 ± 0.20	0.85 ± 0.17	0.40 ± 0.25

F : Flowable composite resin, pior to aging
P : Packable composite resin, pior to aging

The changes of composite surface roughness prior-to and after aging were shown in Figure 2. The graph shows that the roughness changes are likely to decrease after coated with nanofilled self-adhesive protective agent.

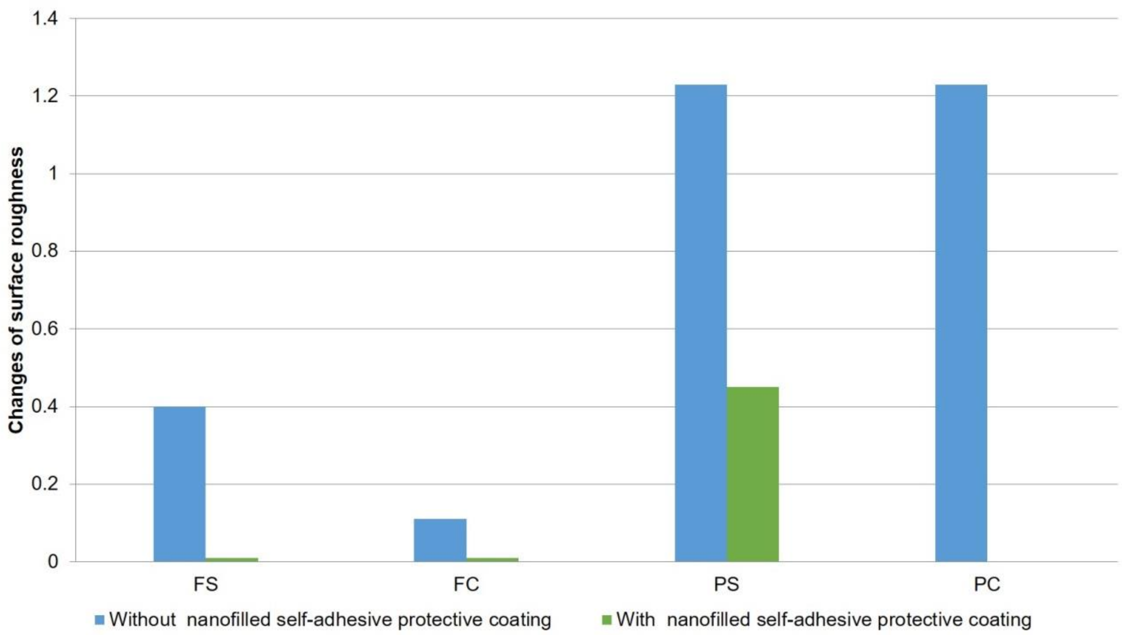

Figure 2. The changes of surface roughness (Ra) values of different composite and aging method.

DISCUSSION

In the current study, the effect of nanofilled self-adhesive protecting coating on the color stability and surface roughness of flowable and packable composite after aging were evaluated. Based on the results obtained, there were significant differences in color changes, and surface roughness of aged flowable and packable composite that were coated with nanofilled self-adhesive protective agent compare to those without coating. It is confirmed that the tested hypothesis was accepted.

Results of the present study showed that aging process under saliva and carbonated drink have adverse effects on changes in ΔE color values of flowable and packable composite, either with or without protective coating. There were published studies that describe the stability of a composite-resin material's color is affected by various chemical factors, such as filler, monomer, activator, pigment, and degree of conversion.[23,24] In this study, artificial saliva and carbonated drink were used to simulate the intra oral aging process. The water-sorption of resin-monomers resulting the different levels of color-stability. As highlighted by Rinastiti, et al.[19] the water lead to softening of the composite-resin surface due to the penetration of water, resulted in hydrolitic degradation of silane that is coated filler particles. Also, water penetration may cause the swelling of the matrix, resulted in the failure of adhesion between filler particle and matrix. This failure will reduce the hardness of composite matrix, whole the appearance of filler particle on the composite-resin's surface resulted the increasing of surface roughnesses. Instead of water effect, a low pH (2.52) of carbonated drink used in the study owing to the swelling of the matrix as well.[25] Moreover, it is also known that the TEGDMA in the matrix of resin composite is hydrophilic thus may increase the water sorbtion and softening the composite surface.[26]

The color changes of flowable composites were founded greater than that packable composite. Analysis of quantitative color values achieved in this study presented that the ΔL, Δa* and Δb*color coordinates of flowable composite was more affected than that packable composite.

This could be due to the composition of packable composite that is contained of UDMA that less hydrophilic compared to TEGDMA. Additionally, the size, distribution, and %w or %v of filler particles may affect the discoloration of the composite-resin.[27] Filler particle loading of flowable composite-resin (47 % vol) is lower than the packable composite-resin (60 % vol). This likely resulted in a greater matrix softening that increase the surface roughness so that the coloring substances contained in saliva and carbonated beverage will be trapped between the uneven surface and increase discoloration of composite-resin.

The surface roughnesses of composite types prior to- and after aging, except fresh flowable composite, were more than 0.02 μm. Previous studies have been reported that a higher surface roughness (> 0.2 μm) exhibit extensive plaque accumulation on dental materials and known as main contributor to the multifactorial discoloration of composite restorations.[28,29] Based on the results, the surface roughness of the aged packable composite was higher than the flowable one. Previous study has confirmed that the smoother surface was correlated with the amount of resin.[30] The filler loading of flowable and packable composite in this study was 47 % vol and 60 % vol respectively.

Application of nanofilled self-adhesive protective coating reducing the color change and surface roughnesses of both composite-resin aged in saliva and carbonated drink. The coating material is composed of monomer and nanofilled particles distributed uniformly in the matrix. The coating on the surface of the composite-resin restoration may enhance degree of polymerization and reduce the degradation due to aging process. The previous study showed that the application of nanofilled self-adhesive protective coating on glass ionomer cement would inhibit the penetration of water better than varnish.[31] The protective coating application may inhibit the penetration of water into the composite, reducing the hydrolysis of the adhesion between matrix and filler particle thereby reducing composite degradation. As a result of those phenomena, composite color was more stable, and the surface was smoother even though the composite has aged. The above finding is consistent with such findings revealed that the thin layer of surface coating material might eliminate the irregular or defects surface of inadequately polished composite restorations.[29]

In principle, after composite-resin is exposed to testing environment, the color difference will not be detected as long as the material is completely color stable or unstained by colorations ($\Delta E = 0$). Nonetheless, from the clinical point of view, when ΔE is >1, the color change is considered detectable to the naked eye, and when ΔE is ≤ 3.3, changes are clinically acceptable.[32] The color changes of composites after protective coating were reduced significantly. Even though the color changes of flowable composite still can be detected to the naked eye, it is clinically acceptable. These findings suggest that in general, the nanofilled self-adhesive protective coating may enhance the color stability of composite restoration. To convince the long-term effect of the protective coating agent, a further study is necessary to evaluate the adhesion between protective coating and the surface of composite-resin. Furthermore the degradation of the coating under various intra-oral conditions should be evaluated.

CONCLUSION

Within the limitations of this study, it can be concluded that the color changes and surface roughness of flowable and packable composite-resin after aging condition were decreased by applying nanofilled self-adhesive protective coating.

ACKNOWLEDGMENTS

This study was supported financially by Faculty of Dentistry, Universitas Gadjah Mada. We are also grateful to Ari Jimi Febriyanto for assistance with roughness measurement.

CONFLICT OF INTEREST

The author reports no conflicts of interest in this work.

REFERENCES

1. Sharif MO, Catleugh M, Merry A, Tickle M, Dunne SM, Brunton P, Aggarwal VR, Chong LY. *Replacement versus repair of defective restorations in adults: resin composite*. Cochrane Database Syst Rev. 2014; Feb 8;2014(2):CD005971.
2. Kanzow P, Krois J, Wiegand A, Schwendicke F. *Long-term treatment costs and cost-effectiveness of restoration repair versus replacement. Dent Mater*. 2021; 37(6):e375-e381.
3. Alawjali SS, Lui JL. *Effect of one-step polishing system on the color stability of nanocomposites*. J Dent. 2013; 41 (Suppl 3):e53-61.
4. Poggio C, Beltrami R, Scribante A, Colombo M, Chiesa M. *Surface discoloration of composite resins: Effects of staining and bleaching*. Dent Res J. 2012 ; Sep;9(5):567-73.
5. Sufyan Garoushi, Lippo Lassila, Marwa Hatem, Muneim Shembesh, Lugane Baady, Ziad Salim & Pekka Vallittu. *Influence of staining solutions and whitening procedures on discoloration of hybrid composite resins*. Acta Odontol Scand. 2013; 71(1):144-150.
6. Bayne SC. *Correlation of clinical performance with 'in vitro tests' of restorative dental materials that use polymer-based matrices*. Dent Mater. 2012; 28(1):52-71.
7. Alshali RZ, Salim NA, Satterthwaite JD, Silikas N.J. *Long-term sorption and solubility of bulk-fill and conventional resin-composites in water and artificial saliva*. J Dent. 2015; 43(12):1511-8.
8. Pilo R, Nissan J, Shafir H, Shapira G, Alter E, Brosh T.J. *The influence of long term water immersion on shear bond strength of amalgam repaired by resin composite and mediated by adhesives or resin modified glass ionomers*. J Dent. 2012; 40(7):594-602.
9. Domingos PA, Garcia PP, Oliveira AL, Palma-Dibb RG. *Composite resin color stability: influence of light sources and immersion media*. J Appl Oral Sci. 2011; 19(3):204-11.
10. Krüger J, Maletz R, Ottl P, Warkentin M. *In vitro aging behavior of dental composites considering the influence of filler content, storage media and incubation time*. PLoS One. 2018 ; Apr 9;13(4):e0195160
11. Rinastiti M, Özcan M, Siswomihardjo W, Busscher HJ. *Effects of surface conditioning on repair bond strengths of non-aged and aged microhybrid, nanohybrid, and nanofilled composite resins*. Clin Oral Investig. 2011;15(5):625-33.
12. Kusuma Yulianto HD, Rinastiti M, Cune MS, de Haan-Visser W, Atema-Smit J, Busscher HJ, van der Mei HC. *Biofilm composition and composite degradation during intra-oral wear*. Dent Mater. 2019; 35(5):740-750.
13. Drumond, J.L. *Degradation, fatigue, and failure of resin dental composite materials*. J Dent Res. 2008; 87(8): 710-719.
14. Attin, T. & Wagehaupt, F.J. *Impact of erosive conditions on tooh-colored restorative materials.* Dent. Mater. 2014; 30(1): 43-49
15. Kafalia, R.F., Firdausy, M.D., Nurhapsari, A. *Pengaruh jus jeruk dan minuman berkarbonasi terhadap kekerasan permukaan resin komposit*. Odonto Dental Journal. 2017; 4(1): 38-43
16. Topcu, F, G. Sahinkesen, K. Yamanel, U. Erdemir, E.A. Oktay & S. Ersahan. *Influence of different drinks on the colour stability of dental resin composites*. Eur J Dent. 2009;3: 50–56.
17. Nasim, I., P. Neelakantan, R. Sujeer, C.V. Subbarao, *Color stability of microfilled, microhybrid and nanocomposite resin - An in vitro study*. J Dent. 2010; 38(S2): e137-3142.
18. Yap, A.U.J., C.W. Sau & K.W. Lye. *Effects of aging on repair bond strengths of a polyacid-modified*. Clin Oral Investig. 2011;15: 625–633.
19. Rinastiti, M., M. Özcan, W. Siswomihardjo, H.J. Busscher & H.C. van der Mei. *Effect of biofilm on the repair bond strengths of composites*. J Dent Res. 2010;89: 1476–1481.
20. Mahmoud, S.H., N.R. El-Kholany, M.E. Grawish & S.A.E. El-Negoly. *In vitro impact of a glaze/composite resin sealant on the surface roughness and bacterial adhesion of one microhybrid and three nanofilled composite resins*. Am J Esthet Dent. 2013;3: 130–142.
21. Kumar, A.A., V.P. Hariharavel, A. Narayanan & S. Murali. *Effect of protective coating on marginal integrity of nanohybrid composite during bleaching with carbamide peroxide: A microleakage study*. Indian J Dent Res. 2015; 26: 167–169.
22. Commision Internationale de L'eclairage. *Recommendations on Uniform Color Spaces, Color Difference Equations, Psychometric Color Terms*. CIE Publisher, Cambridge, 15(Suppl.2), 9–12; 1979.

23. McCabe, J.F. & S. Rusby. *Water absorption, dimensional change and radial pressure in resin matrix dental restorative materials*. Biomaterials. 2004; 25: 4001–4007.
24. Ferracane, J.L. *Hygroscopic and hydrolytic effects in dental polymer networks*. Dent Mater. 2006;22: 211–222.
25. Rahim TN, Mohamad D, Md Akil H, Ab Rahman I. *Water sorption characteristics of restorative dental composites immersed in acidic drinks*. Dent Mater. 2012;28(6):e63-70.
26. Nurhapsari, A. & Kusuma, A.R.P. *Penyerapan air danl kelarutan resin komposit tipe microhybrid, nanohybrid, packable dalam cairan asam.* Odonto Dental Journal. 2018; 5(1): 67-75
27. Barutcigil, C. & M. Yildis*. Intrinsic and extrinsic discoloration of dimethacrylate and silorane based composites*. Journal of Dentistry 40(Supplement 1): e57–e63 (2012).
28. Barakah, H.M. & N.M. Taher. *Effect of polishing systems on stain susceptibility and surface roughness of-ánanocomposite resin material*. J Prosthet Dent.2014; 112: 625–631.
29. Zimmerli, B., T. Koch, S. Flury & A. Lussi. *The influence of toothbrushing and coffee staining on different composite surface coatings*. Clin Oral Investig. 2012;16: 469–479.
30. Bashetty K, Joshi S.J . *The effect of one-step and multi-step polishing systems on surface texture of two different resin composites*. J Conserv Dent. 2010;13(1):34-8.
31. Bayrak, S., T. E Sen, N. Tuloglu & G. Ceylan. *Effects of self-etch adhesives used as surface coating agents on microleakage of conventional and resin modified glass ionomer cement*. Mater Res Innov. 2011;15(1): 53–57.
32. Seghi, R.R., E.R. Hewlett & J. Kim*. Visual and instrumental colorimetric assessments of small color differences on translucent dental porcelain*. J Dent Res.1989; 68: 1760–1764.