

Original Research Article

Effect of four commercial mouth rinses on the microhardness and solubility of a supra nanocomposite and a microhybrid composite: An in vitro study

Dr. Ayush Goyal¹, Dr. John V. George², Dr. Sylvia Mathew³, Dr. Ritu Singh⁴, Dr. Poornima Ramesh⁵

¹Lecturer, Dept. Of Conservative Dentistry & Endodontics, Subharti Dental College, Meerut, Uttar Pradesh, India.

²Professor, Dept. Of Conservative Dentistry & Endodontics, M. S. Ramaiah Dental College, Bengaluru-560094, India

³Professor and HOD, Dept. Of Conservative Dentistry & Endodontics, M. S. Ramaiah Dental College, Bengaluru-560094, India

⁴Lecturer, Dept. Of Paediatric and Preventive Dentistry, Subharti Dental College, Meerut, Uttar Pradesh, India

⁵Lecturer, Dept. Of Conservative Dentistry & Endodontics, M. S. Ramaiah Dental College, Bengaluru-560094, India

***Corresponding author**

Dr. Ayush Goyal

Email: ayush2106_goyal@yahoo.co.in

Abstract: The aim of this study was to compare and evaluate the effect of four commercially available mouth rinses on the microhardness and solubility of a newly introduced resin-composite namely Estelite® α with Filtek™ P60. The microhybrid and the supra-nano composites used in this study are Filtek™ P60 and Estelite® α respectively. A total of 108 (N) specimens (54 for each resin composite) were fabricated and were further divided into 4 subgroups according to the mouth rinse used as follows- subgroup 1- Listerine; subgroup 2- Colgate Plax; subgroup 3-HiOra; sub-group 4- Clohex Plus. Subgroup 5 consisted of the remaining 12 specimens which served as controls (distilled water). Change in the microhardness and solubility of both the resin composites were determined after exposure to all the five test solutions. For microhardness, intra-group comparison showed significant reduction in the microhardness in all the subgroups in both the tested resin-composites ($p < 0.001$). Inter-group comparison showed that the difference in microhardness between the groups was statistically significant only for Colgate Plax ($p < 0.001$) and HiOra ($p < 0.05$). For solubility, maximum solubility was presented by Listerine and minimum by HiOra in both the resin-composites. Inter-group comparison showed that the difference in the solubility of the two resin-composites was statistically significant only for HiOra ($p < 0.05$). Specimens tested in distilled water did not show any significant change for both the tested parameters ($p > 0.05$). Following conclusions can be drawn from the present study- (1) All the mouth rinses used in the study, irrespective of the presence or absence of alcohol reduced the microhardness of both the tested resin-composites. (2) Alcohol content is not the only factor in mouth rinses that can degrade materials. (3) Alcohol-free mouth rinses may be preferable to alcohol containing mouth rinses in patients with extensive restorations.

Keywords: Microhardness, Mouth rinse, Solubility, Composite resin, Supra-nano Composite, Microhybrid Composite

INTRODUCTION

Resin composites are polymer-based materials and hence may undergo degradation in the oral environment [1]. It is reported that saliva, food components and beverages may have an adverse effect on dental composites [2]. Of late, the use of proprietary mouth rinses has become popular [3]. Besides being an effective adjuvant for prevention of caries and gingivitis and as a topical applicant in oral lesions, people tend to use mouth rinses for social and cosmetic reasons [4]. Mouth rinse solutions have various components such as detergents, emulsifiers, organic acids, dyes and alcohol (predominantly ethanol). Alcohol is added to mouth rinses to act as a carrier agent for other active

constituents, to disintegrate plaque and as an antiseptic [5].

In vitro studies have reported that composite resins exposed to ethanol exhibited lower microhardness values compared to non-exposed controls [6]. Also, the degradation effect of alcohol on resin composites has been found to be directly related the percentage of alcohol in the mouth rinses [7]. Excessive solubility of dental restorative materials can lead to marginal discrepancy and hence microleakage. Leakage of fillers, ions, and organic substances such as residual monomers, methacrylate and formaldehyde from resin composite material can occur in the aqueous environment of oral cavity [8].

Previous studies done to evaluate these two properties of composite resins have met with conflicting results [4-5, 9-16]. The aim of this study was to compare and evaluate the effect of four commercially available mouth rinses; Listerine, Colgate Plax, HiOra and Clohex Plus on the microhardness and solubility of

a newly introduced resin-composite namely Estelite®α with Filtek™P60.

MATERIALS & METHODOLOGY

The details of the tested composite resins and the commercial mouth rinses are shown in Tables 1 and 2.

Table 1: Product profile of the tested resin composites used in the study

Material	Specification	Composition			Manufacturer
		Polymeric matrix	Filler Type and load	Filler particle size	
Estelite®α	Supra-nano Composite	Bis-GMA TEGDMA	Silica-Zirconia (82 wt %)	0.1-0.3 µm	Tokuyama, Taitou, Tokyo, Japan
Filtek™P60	Microhybrid Composite	Bis -GMA Bis-EMA UDMA TEGDMA	Zirconia-Silica (78.8 wt %)	0.01-3.5 µm	3M, ESPE, St. Paul, MN, USA

Table 2: The mouth rinse brand name, specification, composition and manufacturer

Mouth rinse	pH	Composition	Manufacturer
Listerine (Alcohol based)	3.72	Purified water, Sorbitol, Alcohol, Poloxamer 407, Benzoic Acid, Sodium Sachharin, Eucalyptol (0.09% w/v), Flavour, Methyl Salicylate, Thymol (0.06% w/v), Sodium Benzoate, Menthol (0.04% w/v), CI 42053	Johnson & Johnson Limited, Bengaluru, India
Colgate Plax (Alcohol free, fluoride-containing)	6.8	Water, Glycerin, Propylene Glycol, Sorbitol, Poloxamer 407, Flavour, Sodium Sachharin, Cetylpyridinium Chloride, Sodium Fluoride, Methylparaben, Phosphoric acid, Menthol, Propylparaben, Camellia Leaf Extract, CI 19140, CI 420512	Colgate-Palmolive Ltd., Thailand
HiOra (Alcohol free, herbal)	4.43	Pilu (Salvadorapersica)- 5mg Nagavalli (Piper betle)- 10 mg Bibhitaka (Terminalibellerica)- 10 mg Peppermint satva (Mentha spp.)- 1.6 mg Yavanisatva (Trachyspermumammi)- 0.4 mg Gandhapurataila (Gaultheria fragrantissima) - 1.2 mg Ela (El. cardamomum) - 0.2 mg	The Himalaya Drug Company, Bangalore, India
Clohex Plus (Alcohol free, CHX containing fluoridated mouth rinse)	5.76	Chlorhexidine Gluconate (0.2% w/v), Sodium Fluoride, Zinc Chloride (0.09% w/v)	Group Pharmaceuticals Ltd, Malur, India

Fabrication of molds:

A total of 108 plastic disks were prepared from a plastic mold and custom modified to get the desired size. Each disk had an external diameter of 8 mm and a thickness of 3.3 mm. The thickness of each plastic disk was verified with a digital calliper (Aerospace Digital Vernier Caliper, India).

Specimen preparation:

Each mold was placed on a microscopic glass slide (Blue Star Frosted Micro Slides, Polar Industrial

Corporation, Mumbai, India). An amount of resin-composite sufficient to slightly overfill the mold was extruded from the tube. The material was then packed in place using a composite placement instrument (Composite filling instrument, GDC, India). Another Mylar strip was placed on the top of the mold and further covered with a second glass slide and pressed for 30 seconds to extrude the excess material and to obtain a uniformly smooth specimen surface.

The mean light intensity of the light source was determined with a commercial radiometer prior to starting the experiment. Each specimen was light cured continuously for 40 seconds from the top then extra 40 seconds from the bottom of the specimen using an LED curing gun (Elipar™ 2500, 3M ESPE Dental products, US) with a light intensity of not less than 450 mW/cm².

The specimens were then immersed in 20 ml of artificial saliva for 24 hours to allow for post-irradiation hardening/post-setting polymerization. At the end of this period, all the specimens were then finished with coarse, medium-coarse, fine and extra-fine Sof-Lex disks (3M, St Paul, MN, USA). Finally, the specimens were polished with felt disks impregnated with light-orange aluminium grit (30-µm slurry; 3M ESPE Dental Products 2385P).

Grouping of the Specimens for Base Line Measurements:

A total of 54 specimens were fabricated for each composite resulting in a total of 108 samples (N). These samples were further subdivided according to the mouth rinse solution in which they had been immersed (n=12). The remaining 12 specimens served as controls. Each specimen group was then immersed in 20 ml of artificial saliva (pH = 7.14) prior to baseline assessment. The specimens were removed from artificial saliva using a tweezer and blotted dry using a filter paper.

Baseline microhardness value:

The baseline microhardness values of the specimens were determined using a Vicker's microhardness tester (HWMMT-X7, Highwood TTS Unlimited, and Japan). Each specimen was positioned centrally beneath the indenter of a digital microhardness tester. A 500g load was applied through the indenter with a dwell time of 15 seconds. Three readings were taken for each specimen and averaged to form a single value (MH₁) for that specimen.

Baseline value for measurement of solubility:

After blotting dry the discs with filter paper, the disks were stored in a glass desiccator at 37°C and weighed on a precision analytical scale (Afcoset Electronic Balance, The Bombay Burmah Trading Company, India) until mass variation was less than ± 0.1 mg.

pH Evaluation:

The pH of the four commercial mouth rinses was recorded using a digital pH meter (Lutron PH-206, PH-MV-temp. Meter, India)

Immersion of the Specimens in the treatment solutions:

Twice a day, the discs were immersed in 20 ml of each mouth rinse for two minutes (12 hour intervals). After immersion in the respective mouth rinses, the

discs were washed in distilled water and kept again in artificial saliva for the remainder of the period.

After treatment measurements:

After seven days, the discs were removed from the vials, washed thoroughly in distilled water and blotted dry with filter paper. The specimens were placed in a glass desiccator and weighed until the mass variation was less than ± 0.1 mg (m₂). The solubility was obtained using the following formula:

$$SI = \frac{m_1 - m_2}{V}$$

Where,

SI = Solubility

m₁ = Pre-treatment mass after drying (mg)

m₂ = Post-treatment mass after drying (mg)

V = Volume in mm³

The specimens were then subjected to microhardness test and the post-treatment values (MH₂) determined. The change in hardness value between the baseline and after treatment measurement was calculated according to the following equation:

$$\Delta VHN = MH_1 (\text{baseline}) - MH_2 (\text{after treatment})$$

STATISTICAL ANALYSIS:

2-way ANOVA was used to compare the intra-group change in microhardness presented by the four mouth rinses. One-way ANOVA with Post-Hoc Scheffe test was used to compare the inter-group change in microhardness and mean difference in solubility between the two resin-composites.

RESULTS

Table 3 illustrates the effect of five test solutions on the tested resin composites. Intra-group comparison showed significant reduction in the microhardness in all the subgroups after immersion in the mouth rinses compared to baseline values in both the tested resin-composites (p < 0.001). The maximum reduction in microhardness was presented by Listerine in both tested resin-composites (p < 0.001). HiOra produced the lowest changes in the microhardness of Estelite®α (p < 0.001) and Filtek™P60 (p < 0.05) followed by Clohex Plus, Colgate Plax, and Listerine. Inter-group comparison showed that the difference in microhardness between the groups was statistically significant only for Colgate Plax (p < 0.001) and HiOra (p < 0.05). With regard to solubility, similar results were obtained as for microhardness i.e. maximum solubility was presented by Listerine and minimum by HiOra in both the resin-composites. Inter-group comparison showed that the difference in the solubility of the two resin-composites was statistically significant only for HiOra (p < 0.05). Upon exposure to distilled

water, there was negligible or no change in the pre-treatment values for both the groups (for mean

difference in microhardness as well as solubility.

Table 3: Comparison of mean difference in microhardness and solubility in various mouth rinses

No.	Mouth rinse	Mean Difference in microhardness (MH ₁ -MH ₂) Mean ± SD		Solubility Mean ±SD	
		Estelite® α	Filtek™P60	Estelite® α	Filtek™P60
1	Listerine	5.86 ± 2.89 ^{Aa}	7.03 ± 1.46 ^{Aa}	7.04 ± 0.64 ^{Aa}	7.3 ± 1.20 ^{Aa}
2	Colgate Plax	2.80 ± 1.18 ^{Ab}	2.64 ± 1.00 ^{Bb}	6.83 ± .88 ^{Aa}	6.16 ± 1.10 ^{Ab}
3	HiOra	1.40 ± 0.92 ^{Ab}	0.35 ± 0.33 ^{Bc}	5.32 ± 0.40 ^{Ab}	4.97 ± .40 ^{Bc}
4	Clohex Plus	2.10 ± 0.82 ^{Ab}	2.01 ± 0.83 ^{Abd}	6.35 ± .76 ^{Aa}	6.11 ± 0.99 ^{Ab}
5	Distilled water	0.08 ± 0.05 ^{Ac}	0.11 ± 0.04 ^{Ac}	0.56 ± 0.02 ^{Ac}	0.45 ± 0.05 ^{Ad}

Means followed by different superscript letters (capital letters in row and lower case letters in column) indicate statistically significant difference ($p < 0.05$) and same superscript letters indicate no significant difference.

DISCUSSION

Literature on the effect of mouth rinses on the microhardness and solubility of newly introduced composites is limited, hence this motivated us to fill this lacuna in literature and to conduct the present study using commonly used mouth rinses in the Indian market.

Estelite® α , a supra-nano spherical filled resin composite, has been recently introduced in the market with inadequate literature to support its clinical use. Estelite® α was developed based on the sol-gel method that controls the diameter of fillers and changes the refractive index of the fillers. Data from the manufacturer shows that this material has superior mechanical properties owing to its unique manufacturing process.

Hardness is included as one of the test parameters, as it is an important property for the restorative materials to have long-term durability in the oral cavity [17]. Long term service cannot be expected from a restorative material which is susceptible to chemical degradation on exposure to mouth rinses.

Previous studies have reported that the maximum change in the hardness of composites occurred within the first 7 days after exposure to food simulating liquids [18]. For this reason, the specimens in this study were immersed in the respective mouth rinses for 1 week before undertaking the microhardness and solubility tests.

In our study, all the mouth rinses irrespective of the presence or absence of alcohol resulted in significant reduction in the microhardness of both the tested resin composite materials compared to the base line values. This may be attributed to the acidic pH of mouth rinses which would have resulted in erosion of the resin composites by acid etching and leaching the principle matrix forming cations. Another explanation for this phenomenon is catalysis of ester groups in

dimethacrylate monomers present in the organic matrix (Bis-GMA, Bis-EMA, UDMA and TEGDMA) due to their acidic nature [5]. Hydrolysis of these ester groups forms alcohol and carboxylic acid as by-products which may further accelerate the degradation process. Our results are not entirely in agreement with the observations by Diab et al in 2007 who reported that mouth rinses with low pH are deleterious to the hardness of resin composites [14]. Acidic pH might cause some erosion of the composite materials as mentioned above but it cannot be the sole cause of deterioration. In the present study, statistically significant difference was observed for Colgate Plax (pH =6.8) and HiOra (pH=4.43) with regard to microhardness.

The effect of mouth rinses on hardness is dependent on the chemical composition of the restorative material. The hydrophilicity of various matrix monomers follows the order: TEGDMA>Bis-GMA>UDMA>HMDMA [19]. Thus, TEGDMA is more susceptible to enzymatic hydrolysis than Bis-GMA or Bis-EMA. Estelite® α primarily consists of Bis-GMA and TEGDMA as its polymeric matrix and this explains the significant reduction in microhardness caused by Listerine.

A dental composite consists of different types of inorganic fillers. Estelite® α has a mean filler particle size of 0.2 μm whereas mean particle size of the fillers present in Filtek™P60 is 0.6 μm . Though mean particle size of Estelite® α is smaller than Filtek™P60, but latter incorporates a larger volume fraction of smaller filler particles [20] (Table 1). In addition to Bis-GMA, the matrix of this composite resin contains UDMA and Bis-EMA. Also, to incorporate higher volume of fillers in the polymeric matrix, Filtek™P60 has 60% of small silica particles [21].

The aforementioned factors could have resulted in slightly better performance of Filtek™P60. Apart from filler size, the type of filler also has an

effect on dissolution dynamics. Composites containing zinc and barium glass fillers have been shown to be more susceptible to aqueous attack than those containing quartz filler [22]. Also, Yap *et al.* observed that zirconia glass fillers were susceptible to aqueous attack [23]. In the present study, both Estelite® α and Filtek™P60 contained synthetic zirconia/silica fillers and this may explain their inferior performance against all the mouth rinses.

Solubility mean values presented by the composite resins tested varied from 4.97 to 7.3 $\mu\text{g}/\text{mm}^3$. These values were lower than the maximum value certified by the ISO 4049 standard ($<7.5 \mu\text{g}/\text{mm}^3$) [8]. Although direct extrapolation is not possible, the results of the present study showed that the solubility of all materials in all solutions is acceptable for the ISO 4049 standard.

In the present study, although immersion in Listerine caused maximum values of solubility, the difference between both the tested resin-composites was not statistically significant. On the other hand, minimum values of solubility were seen on immersion in HiOra and this difference was statistically significant between both composite restorative materials.

Negligible change in the values for microhardness and solubility was seen under the influence of distilled water for both the groups. Therefore, it can be theorized, that interaction of chemical components present in mouth rinses and their acidic pH influences the mechanical properties of resin composites. Also, alcohol influences the hardness and solubility of composite resins, but the effect of alcohol does not take place by its own and hence, there must be a simultaneous interaction of other factors that affect the physical properties of composite resins.

In the present study, both Estelite® α and Filtek™P60 exhibited greater solubility in Listerine (alcohol 21.6% w/v) when compared with other alcohol-free mouth rinses. Thus, it is recommended for patients with extensive restorations to avoid the use of mouth rinses containing alcohol as a routine practice. As with all *in vitro* studies, caution must be used when extrapolating the results of the present study to the oral environment.

CONCLUSION

Within the limitations of the experimental design and the test parameters, it can be concluded that-

1. All the mouth rinses used in the study, irrespective of the presence or absence of alcohol reduced the microhardness of both the tested resin-composites.
2. Solubility and reduction in microhardness, values of both the tested parameters were lower in alcohol-free mouth rinses than in Listerine.

3. Amongst all the alcohol-free mouth rinses HiOra caused least reduction in microhardness and resulted in least solubility of both the tested resin-composites.
4. Alcohol content is not the only factor in mouth rinses that can degrade materials.
5. Alcohol-free mouth rinses may be preferable to alcohol containing mouth rinses in patients with extensive restorations.

REFERENCES

1. Ferracane JL. Hygroscopic and hydrolytic effects in dental polymer networks. *Dental Materials*. 2006 Mar 31; 22(3):211-22.
2. Lee SY, Huang HM, Lin CY, Shih YH. Leached components from dental composites in oral simulating fluids and the resultant composite strengths. *Journal of Oral Rehabilitation*. 1998 Aug 1; 25:575-88.
3. Walsh TF. Mouthrinses as adjuncts in periodontal therapy. *Dental update*. 1996 May; 23(4):144-7.
4. Gürdal P, Akdeniz BG, Hakan Sen B. The effects of mouthrinses on microhardness and colour stability of aesthetic restorative materials. *Journal of oral rehabilitation*. 2002 Sep 1; 29(9):895-901.
5. Yap AU, Tan BW, Tay LC, Chang KM, Loy TK, Mok BY. Effect of mouthrinses on microhardness and wear of composite and compomer restoratives. *Operative dentistry*. 2002 Dec; 28(6):740-6.
6. Schneider LF, Moraes RR, Cavalcante LM, Sinhoreti MA, Correr-Sobrinho L, Consani S. Cross-link density evaluation through softening tests: effect of ethanol concentration. *dental materials*. 2008 Feb 29; 24(2):199-203.
7. Penugonda B, Settembrini L, Scherer W, Hittelman E, Strassler H. Alcohol-containing mouthwashes: effect on composite hardness. *The Journal of clinical dentistry*. 1993 Dec; 5(2):60-2.
8. Ozer S, Sen Tunc E, Tuloglu N, Bayrak S. Solubility of Two Resin Composites in Different Mouthrinses. *BioMed research international*. 2014 Apr 7; 2014.
9. Almeida GS, Poskus LT, Guimarães JG, Silva EM. The effect of mouthrinses on salivary sorption, solubility and surface degradation of a nanofilled and a hybrid resin composite. *Operative dentistry*. 2010 Jan; 35(1):105-11.
10. Gürkan S, Önen A, Köprülü H. *In vitro* effects of alcohol-containing and alcohol-free mouthrinses on microhardness of some restorative materials. *Journal of oral rehabilitation*. 1997 Mar 1; 24(3):244-6.
11. Jyothi KN, Crasta S, Venugopal P. Effect of five commercial mouth rinses on the microhardness of a nanofilled resin composite restorative

- material: An in vitro study. Journal of conservative dentistry: JCD. 2012 Jul; 15(3):214.
12. Festuccia MS, Garcia LD, Cruvinel DR, Pires-De-Souza FD. Color stability, surface roughness and microhardness of composites submitted to mouth rinsing action. Journal of Applied Oral Science. 2012 Apr; 20(2):200-5.
 13. De Moraes PI, das Neves LE, de Souza CK, Parolia A, Barbosa DS. A comparative effect of mouthwashes with different alcohol concentrations on surface hardness, sorption and solubility of composite resins. Oral health and dental management. 2014 Jun; 13(2):502-6.
 14. Diab M, Zaazou MH, Mubarak EH, Olaa MI. Effect of five commercial mouthrinses on the microhardness and color stability of two resin composite restorative materials. Aust J Basic Appl Sci. 2007; 1(4):667-74.
 15. Miranda DD, Bertoldo CE, Aguiar FH, Lima DA, Lovadino JR. Effects of mouthwashes on Knoop hardness and surface roughness of dental composites after different immersion times. Brazilian oral research. 2011 Apr; 25(2):168-73.
 16. Fernandez RA, El Araby M, Sibli M, Al-Shehri A. The effect of different types of oral mouth rinses on the hardness of Silorane-based and Nano-hybrid composites. Saudi Journal of Oral Sciences. 2014 Jul 1; 1(2):105.
 17. Okada K, Tosaki S, Hirota K, Hume WR. Surface hardness change of restorative filling materials stored in saliva. Dental Materials. 2001 Jan 31; 17(1):34-9.
 18. Kao EC. Influence of food-simulating solvents on resin composites and glass-ionomer restorative cement. Dental Materials. 1989 May 31; 5(3):201-8.
 19. Venz S, Dickens B. NIR-spectroscopic investigation of water sorption characteristics of dental resins and composites. Journal of biomedical materials research. 1991 Oct 1; 25(10):1231-48.
 20. Murchison DF, Moore BK. Influence of curing time and distance on microhardness of eight light-cured liners. Operative dentistry. 1991 Dec; 17(4):135-41.
 21. Della Bona Á, Benetti P, Borba M, Cecchetti D. Flexural and diametral tensile strength of composite resins. Brazilian oral research. 2008 Mar; 22(1):84-9.
 22. Soderholm KJ. Leaking of fillers in dental composites. Journal of Dental Research. 1983 Feb 1; 62(2):126-30.
 23. Yap AU, Tan SH, Wee SS, Lee CW, Lim EL, Zeng KY. Chemical degradation of composite restoratives. J Oral Rehabil, 2011; 28: 1015-21.