

CHAPTER 5
DISCUSSION

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Soderholm, (1981) studied the degradation of glass-filled experimental composites by measuring pH and sodium content after immersion in water for 16, 32 and 80 days. He found that pH of water increases with time and sodium ion concentration was highest after 80 days. He then concluded that glass particles could have leached from the experimental composite. This was one of the earliest studies to illustrate surface degradation of composite.

Surface degradation can also be related to softening, where surface hardness is measured. The measurement of surface microhardness of composite is an indirect method of evaluating the mechanical strength, resistance to intraoral softening and degradability of resin-based restorative materials by water and chemicals in the oral environment. This mode of testing has been used by Söderholm, (1981), Wu and McKinney, (1982) and Yap *et al.*, (2001) for measurement of surface degradation of composites due to softening. Water or other chemicals available in the oral cavity could with time, cause filler-matrix debonding, thereby reducing the wear resistance and decreasing the mechanical properties of composites.

In this study, a standardized method was utilized when preparing the specimens to ensure consistency. A Perspex split mould was used for easy removal of the specimens. The matrix strip and glass slide were placed on the mould to ensure adequate and accurate placement of the resin composites into the mould. The matrix strip serves two purposes: first, covering the surface with a non-porous strip will eliminate the oxygen inhibition layer; and secondly, the matrix strip produces a very smooth, regularly contoured surface. However, the polymerized surface against the matrix strip produced

a resin-riched layer, which is less resistant to abrasion and contain air voids (Setcos *et al.*, 1999). This resin-riched layer undergoes wear in the oral environment resulting in exposure of filler particles (Dennison *et al.*, 1981). Therefore, it is desirable that the resin-riched surface be removed. The removal of this layer causes reduction in surface smoothness and it will necessitate polishing (Yap *et al.*, 1998).

Specimens were polished using Sof-Lex polishing discs (3M ESPE Dental St.Paul, Minnesota) as per manufacturer instruction. Hondrum and Fernandez, (1997) showed that the surface of composite specimens polished using Sof-Lex disc produced surface smoothness which was consistently similar to that produced by matrix strip. Kanter *et al.*, (1983) demonstrated that the smoothest surface of composite resin restoration could be produced by using a series of four Sof-lex polishing discs used sequentially from coarse to superfine. Sof-Lex has also been shown to produce the smoothest surface for some composites, such as Esthet X (Dentsply), Venus (Heraeus Kulzer, Dormagen, Germany), InTen-S (Ivoclar Vivadent) and Point B (Kerr), (Gedik *et al.*, 2005). This finding was further substantiated by Mahmood and Lui (2005), where Esthet-X (Dentsply), TPH Spectrum (Dentsply) and Filtek Supreme (3M) were found to have the lowest surface roughness when polished with Sof-Lex.

The consistency in specimen preparation, indentation and measurement of surface microhardness was clearly shown by the result of the control group. Initial VHN of each type of composite serves as the control group to be compared to the VHN after immersion in the five different mouthrinses (Listerine, Oral B and experimental plant extracts X, Y, Z) and distilled water. For each specimen the VHN recorded was a mean of three indentations. One-way ANOVA showed that there was no significant difference between all 60 specimens of each composite except for Ceram•X Mono. Exploration of

data with SPSS Ver.13 revealed that there were three outliers in the data set. Outliers may be removed for statistical procedure as suggested by Tabachnick and Fidell, (2001). Once the outliers were removed, there was no significant difference in the VHN of all 57 specimens. This clearly showed the consistency of specimen preparation of reproducibility of VHN measured. The standard deviation of all measurements was also low (Appendix III). The data set for each composite immersed in the five mouthrinses and distilled water was ten. Prior to immersion, the initial VHN was measured. Therefore, sixty data sets was obtained for each composite. Randomization of these data was necessary in order to select ten dataset for the control group (Appendix III).

Among the composites tested, Filtek Z350 was the hardest (92.6 VHN) compared to other composites. It may be due to the variation in filler type, shape of fillers, percentage of filler loading and whether the fillers are distributed in clusters or as individual particle. The manufacturer claimed that “Filtek Z350 contain a unique combination of individual nanoparticles and nanoclusters. The nanoparticles consisted of nanosilica fillers, which are discrete nano-agglomerated and nano-aggregated particles of 20nm in size. Nanocluster fillers are zirconia/silica and loosely bound agglomerates of nano-sized particles. These agglomerates act as single units enabling high filler loading and high strength” (Technical Product Profile, Filtek, 3M ESPE). It was also suggested that the combination of nano-sized particles to form nanoclusters reduces the interstitial spacing of filler particles thus increasing filler loading (78.5%) and improved hardness. Sukumaran *et al.*, (2005) found that the hardness of Filtek Supreme (enamel shade) was 74.8 VHN. Filtek Z350 contains the same patented and proprietary nanotechnology used to create Filtek™ Supreme as stated by the manufacturer. The resin properties such as handling, strength, polish retention and polymerization shrinkage are unchanged except for various function such as enamel,

dentine (body) and opaque resin. However, Filtek Z350 is being marketed as a composite restorative material without the option of enamel, dentine (body) and opaque resin. The difference in VHN is likely to be due to enamel shade that was used in the earlier study.

Although Ceram•X Mono and Ceram•X Duo-enamel shade are nano-based materials, their hardness were significantly lower compared to Filtek Z350, 79.4 VHN and 72.8VHN respectively. There was no significant difference in the hardness of both materials as they may have the same composition and same types of fillers. Their fillers are barium-aluminium-borosilicate glass and methacrylate functionalised silicon dioxide nano filler (Ceram•X Scientific Compendium, 2003). This could account for the difference in hardness between the Ceram•X composites and Filtek Z350.

Sukumaran *et al.*, (2005) reported that the VHN of Ceram•X Duo enamel shade was 65.0 before immersion in different types of acidic soft drinks. In this study VHN of Ceram•X Duo-enamel shade was 72.8. The higher VHN observed may be due to differences in samples preparation, the intensity of light curing unit, the load applied, the dimension of indentation measurement and materials batch number. These same reasons may be applied to the higher VHN of TPH Spectrum (73.1) observed in this study compared to that reported by Coffey *et al.*, (2004), 61.0 VHN after 24 hours post-radiation with a quartz-halogen lamp and Ray *et al.*, (2006), 62.7 VHN cured by LED light unit for 40 seconds.

The mean VHN of all tested composites after immersion was significantly reduced for all groups compared to VHN before immersion. This reduction in VHN of composites concurred with observation from earlier studies. Wu and McKinney (1982), reported

that chemicals such as ethanol and cyclohexanone can soften the Bis-GMA copolymer and significantly can cause loss in the wear resistance of composite based on this copolymer. Higher wear rate was observed for ethanol-soaked specimens compared to water. The specimens treated with 75/25 ethanol/water mixture has higher initial wear rate, but wear rate tend to plateau over time. The long-term steady-state wear rate probably depend on the rate of desorption and sorption of the chemicals in addition to the wear resistance of the chemically softened structure.

In another study, Asmussen (1983) found that not only ethanol but acetic and propionic acid have softening effect on Bis-GMA based polymers. The softening effect decreases with increasing content of TEGDMA up to 50mol % and is small for contents of TEGDMA above that limit. On the other hand, lactic acid and water had no influence on surface hardness of the composites tested. Acetic and propionic acid may have solubility parameter in the range that can damage the surface of composite but not lactic acid and water.

McKinney and Wu (1985) found that 75% ethanol in water (solubility value of approximately $3 \times 10^4 \text{J}^{1/2} \text{m}^{-3/2}$) caused the most reduction in hardness of conventional and microfilled composites. They implied that any oral and food-ingredient component having a solubility parameter approximating to this value might produce damage in Bis-GMA based composites. The extent of damage may depend on diffusion rate, which, in turn depend on the molecular weight of the penetrant. The damage mechanism was attributed to softening of the polymer matrix, which result in partial removal of filler particles at the surface.

Later in 2001, Yap *et.al* found that the thickest degradation layer was observed after conditioning of Silux in heptane, an organic solvent that has the potential to damage polymer like ethanol/water solution. They also found that material with zirconia glass fillers such as Z100 were also susceptible to aqueous attack. Both organic solvent and water/weak acids may degrade composite resins; however, it was concluded that the effect of chemical medium on hardness and degradation layer thickness was material dependent.

In this present study, univariate analysis of variance revealed that there was significant interaction between composites and mouthrinses including distilled water. Post-hoc test (Games-Howell) later illustrate that the decrease in VHN was material dependent. Although this observation concurred with that of Yap *et al.*, (2003), it was felt that the immersion duration may not be adequate or long enough to elucidate the effect of chemicals in the mouthrinses. This assumption is made based on the non-significant difference between the mouthrinses and distilled water. It is likely that the decrease in VHN was due to water absorption. Water sorption may affect composite materials by reducing the wear resistance (Söderholm, 1981) due to leaching out of ions when stored in water. The leaching process could cause filler-matrix debonding and cause a weakened filler surface. Leaching increases with time indicating that water diffuses through the resin and reacts with the fillers at the filler-matrix interface. It was also found that the leaching process might originate from the polished surface and from hydrolytic degradation of coupling agent (Söderholm, 1984).

The hydrolytic effect of water on surface hardness of composite has been discussed by Milleding *et al.*, 1998, who found that water significantly reduce the microhardness of the studied composites (Variolink-Ivoclar- high and low viscosity) at every test

occasion (1,7 and 60 days) as compared to dry storage conditions. The VHN of specimens stored exhibit lower values compared to baseline (before storage), indicating that water had a pronounced influence on the surface of composites. Composite resin absorbs a small percentage of water, which will affect the surface hardness. The amount of water sorption and the length of storage time both initiate a hygroscopic swelling of the matrix in the surface layer. However, no significance different in water absorption was found between the two viscosities (Variolink-Ivoclar- high and low viscosity) of composites . Therefore, it was concluded that not only the amount of water absorbed and time of exposure affect the hardness but also the effect of water resulting from the composition and microstructure of the composite resin material will influence the microhardness.

Martos *et al.*, (2003), later found that all tested composites stored in distilled water suffered a reduction in surface microhardness. All water-stored specimens showed a significant drop of Knoop microhardness to the dry-stored control. Decreased in Knoop microhardness was found in composites stored in water for 30 days.

Further studies will have to be carried out to substantiate the hydrolytic degradation by water in the mouthrinses in this study. The 24 hours immersion time that was used in this study was an accordance to Yap *et al.*, (2003). However, on hindsight a shorter immersion may be able to elucidate the effect of chemicals in mouthrinses as carried out by Penugonda *et al.*, (1994) where short immersion period daily over a period of time was used. Moreover, once the immersion period was completed, specimens were dried and stored in airtight containers. They reported that different concentrations of alcohol-containing mouthrinses could affect the surface hardness of all tested composites after

six months repeated immersion (two minutes daily). Through this treatment regime, they were able to illustrate the effect of alcohol on surface degradation.

In the present study, the effect of alcohol mouthrinses on surface microhardness was not significantly different from alcohol-free mouthrinses, experimental plant extract mouthrinses and distilled water. The types of mouthrinses did not have an effect the surface microhardness of tested composites. Previous studies have shown that alcohol-containing products can affect selected physical properties (diametral tensile strength) of composite restorative materials such as Z100[®], Heliomolar[®] and Marathon One[®]. (Lee *et al.*, 1998).

Gurgan *et al.*, (1997) reported that both alcohol containing and no-alcohol mouthrinses affect the microhardness of restorative materials compared to distilled water. The differences between microhardness of alcohol containing and no-alcohol mouthrinses were not significant. Gurdal *et al.*, (2002) found that mouthrinses with various pH and alcohol contents has no significant effect on the microhardness and colour stability of all restorative materials tested. They suggested that the difference observed might have originated from the structure of materials themselves, not from test solution. However, this suggestion was not clearly explained.

Even though, post-hoc test showed that the mean VHN were not significantly different between each group of mouthrinses but all three types of experimental plant extract had some effect on Ceram•X Mono where the VHN were lowered. The mean VHN for Ceram•X Mono immersed in all three types of experimental plant extract mouthrinses were low compared to mean VHN of composites in other mouthrinses. The effect on surface microhardness of Ceram•X Mono may be due to the composition of plant

extract because there are no other chemicals in the experimental mouthrinses. The plant extract used in the experimental mouthrinses were mango leaves, guava leaves and mint leaves. All these leaves seem to have phenolic compounds that may have some effect on hardness on Ceram•X composites. Yap *et al.*, (2003) found that the tested composites (Estet X and Dyract) were significantly softened by the alcohol-containing essential oil/phenolic compound mouthrinses Listerine. Mango leaves contains small proportion of phenolic (Lee *et al.*, 1987) and guava leaves also has phenolic compound (Abreu *et al.*, 2006) that may has some effect on the Ceram•X Mono. Phenolic compound was also found in free soluble form and cell wall bound from extracted mint leaves (Dudai, *et al.*, 2006). The antimicrobial, antiplaque and antigingivitis effect of this experimental mouthrinses had been studied before but not the effect on mechanical properties of restorative materials.

Ceram•X Duo-enamel shade was not affected by the experimental plant extract mouthrinses and this may be due to the unique nano-ceramic matrix combination with the optimized filler particle size to create an ideal balance between handling and optical characteristic that provide the translucency of natural enamel (Dentsply Scientific Compendium, 2003).

The experimental mouthrinses contain purely plant extract similarly to herbal mouthrinses available in the market but the types of plant extract are different. Herbal mouthrinses (Tooth and Gum Tonic) contain mainly pure essential oils from red thyme, eucalyptus, peppermint, cinnamon, bark and lavender. This group of products has the ability to act as a protective therapeutic treatment as well a reactive care solution. They have been used as an antimicrobial and for tissue conditioning, connective tissue rebuilding and anti-halitosis. The other compositions are herbal extract of echinacea,

gotu, kola and green tea. These extracts are effective in the maintenance and health of periodontium and dentition (Shuman, 2003).

Surface analysis

Hydrolytic and chemical degradation of the polymer matrix leaving partially exposed filler particles revealed an increased surface roughness of the composites (Larsen and Munksgaard, 1991). In this present study, the surface analysis of composites using AFM was done to see the effect of various mouthrinses. AFM was chosen because it is the recent development in analysing the surface of flat specimens. It provides the 3D topography of the surface and the mean deviation of surface roughness reading in Ra. The average surface mean height deviation value (Ra) is commonly referred to as the surface finish and the most commonly reported parameter for calculation of surface roughness.

Atomic Force Microscopy qualitative analysis of representative samples confirmed the differences among the surface after immersion in different mouthrinses. Only one AFM scanning area (40 μ m x 40 μ m) of the surface of each specimen was done. The scanning result may not represent the surface analysis for entire specimens. Filtek Z350 in Listerine showed the highest Ra (574.5nm) among all the specimens tested. The alcohol content in Listerine may have some effect on Filtek Z350 compared to other composites. In addition, Martos *et al.*, (2003) reported that the SEM evaluation of Z250 stored dry and specimens kept in an aqueous environment revealed changes in surface texture. The wet-stored samples were significantly rougher than the dry-stored specimens were and showed a fine highly porous structure. This surface roughness appeared to be a discernible loss of material and crack formation. Filtek Z250, which has zirconia silicate fillers, showed large softening effect and susceptible to aqueous

attack. They attributed this to the smaller filler surface area associated with the spherical shape of zirconia /silica fillers that may decrease bonding of fillers and resin matrix.

Other composites immersed in Listerine had a comparable surface roughness to each other. This result concurred with Yap *et al.*, (2000) which concluded that the surface roughness of the composites and polyacid-modified composites evaluated was not significantly affected by conditioning in any of food stimulating liquids. However, a large increase in surface roughness was noted for Dyract AP after conditioning in citric acid and ethanol-water solution.

Milleding *et al.*, 1998 found that surface of tested specimens (Variolink-Ivoclar- high and low viscosity) analyzed with confocal laser profilometry showed very small changes in surface roughness, even after water-storage period of 60 days. However, test performed with optical profilometer revealed that the mean height deviation value (Ra) was significantly higher after storage in water for 60 days compared to the baseline (dry).

Turssi *et al.*, (2002) found that all investigated restorative materials (Durafill VS, Filtek Z350, Dyract AP and Fuji II LC) became significantly rougher after immersion in distilled deionised water, artificial saliva and subjected to pH cycling regimen. The lowest surface roughness was measured for the microfilled composite resin. The hybrid composite showed lower surface roughness than polyacid-modified composite resin. Based on environmental scanning electron microscope (ESEM), the surface of microfilled composite resin revealed protruding particles probably because of matrix degradation. It was also found that the matrix of hybrid composite and polyacid-modified composite resin showed particle voids, which might be attributed to

degradation of the surrounding resin matrix or silane-coupling agent. Matrix dissolution of resin-modified glass ionomer cement detected at the periphery of glass particles could be the result of dissolution of the siliceous hydrogel layer. They concluded that resin-based restoratives underwent greater micromorphological damages following the regimen of acid challenge than after storage in either distilled deionised water or artificial saliva.

All the three types of experimental mouthrinses seem to show some effect on Ceram•X Mono because the results showed high Ra and low VHN. No study had been done to evaluate the effect of herbal mouthrinses on mechanical and physical properties of composite restorative materials for comparison with the result from this study.

CLINICAL IMPLICATION

The significant reduction of microhardness of tested composites after immersion in various mouthrinses and distilled water revealed that composites softened after exposure to various chemicals. However, there was no significant difference between types of mouthrinses and distilled water. The decrease in VHN was likely due to water absorption. Even though the reduction of VHN may be due to water only but repeated used of mouthrinses may have an immediate effect that may lead to a loss of marginal integrity and will lead to marginal staining in addition to increased wear rate. In the clinical situation, composite restorative materials in the oral cavity will be exposed to chemicals in saliva and other solutions in the oral environment that may degrade, soften and increase the roughness and wear of the composites. The surface layer that is softened by the chemicals usually will be removed mechanically. The removal of this layer will expose a new surface layer to further chemical attack and thus contribute to the in vivo wear of composite restorations.

LIMITATION OF STUDY

There are a few limiting factors which need to be considered in this study before the results can be further evaluated. The immersion time used in this study was 24hours that was an accordance to Yap *et al.*, (2003) may be too short or too long to illustrate the actual effect of chemicals in mouthrinses.

The detailed components and percentage of plant extracts in experimental mouthrinses were not known due to patent application process. Therefore, which chemical components that has an effect on the tested composites especially Ceram•X Mono, could not be confirmed.

In the methodology of this study, the specimens were immersed in mouthrinses without shaking and vibration during the immersion period. Shaking and vibrating repeatedly will simulate the rinsing procedure and may give the true effect of chemicals in mouthrinses.

Surface roughness of composites prior to immersion was not measured using AFM due to equipment unavailability when the study commenced. It would have been interesting to compare the initial surface roughness to that after immersion in the various mouthrinses.

RECOMMENDATION FOR FURTHER STUDIES

1. The same methodology may be used but with a shorter duration of immersion (two minutes repeated daily) to simulate rinsing procedure.
2. The specimens will be stored dry in airtight containers between immersions compared to wet storage.
3. Chemical analysis of experimental mouthrinses in order to determine the composition and percentage of essential oil and phenolic content.
4. To determine the wear rate of tested composites after exposure to various mouthrinses.