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Effect of Citric Acid on Microhardness and Surface Roughness of Different Composite Resin Formulations

Mona A. Mohammed, Ahmed A. Laithy**, Omaima H. Ghallab****

Abstract:

Objective: This is an in vitro study that was done to evaluate the effect of citric acid of different pH values on the surface characteristics of two resin composite formulations.

Material and Methods: One conventional composite (Filtek Z350 nanofill [NF]) and a bulk-fill composite (Filtek Bulk-fill [BF] were used in the study. A total of 168 disk specimens (5 mm × 2 mm) were fabricated 84 for each type of composite material. Distilled water with a neutral pH (6.5-7) was used as the immersion medium for control groups, while Citric acid solutions with pH values of 2.5 and 5.5 were prepared and used as the immersion media for test groups. Samples were immersed in immersion solutions for either three days or one week. Microhardness and surface roughness were then recorded and collected data were tabulated and statistically analyzed.

Results: Significant differences in Surface characteristics were observed. BF Samples showed better results against acid attack in terms of Microhardness and surface roughness than NF samples.

Conclusions: Decreased pH values of citric acid negatively affected surface characteristics of dental composite restorations.

Keywords: Composites, Citric acid, Surface characteristics.

** Master's Degree student of Operative Dentistry, Faculty of Dentistry, Ain Shams University.*
*** Lecturer of Operative Dentistry, Faculty of Dentistry, Ain Shams University.*
**** Professor of operative dentistry department Faculty of Dentistry, Ain Shams University.*

Introduction:

Resin-based composites are becoming more popular in restorative dentistry, particularly because of their superior esthetic outcomes. They typically consist of a methacrylate based resin matrix (mass fraction of about 25–30%), glass or ceramic fillers (mass fraction of about 70–75%), and filler-matrix coupling agent [1] .The clinical success of an aesthetic direct restorative material depends upon its mechanical behavior, physical and chemical characteristics, as well as clinical indications [2]**.** Currently, Nanofilled resin composite and bulk filled resin composite are the most commonly used materials nowadays due to the tremendous developments that took place in the recent years.

One of the most important properties that determine the durability of dental materials in the oral cavity is their resistance to dissolution or disintegration[3]. Acid erosion has a clinical significance because acidic conditions can occur orally either due to the ingestion of acidic foods or the degradation of poly saccharides to acids in stagnant areas of the mouth[3]. Dietary awareness is an important issue in modern society. The consumption of carbonated drinks is popular with the youth of today and the habit is carried into adulthood. Healthy diets such as fruits, fruit juices and yogurt may as well cause erosion by their acidity [4]. The consumption of acidic foodstuff and beverages plays a major role in the development of erosion. Along with the change of lifestyle through the decades, the total amount and frequency of consumption of acidic foods and drinks have also changed. The potential erosive effect of a soft drink depends on a number of conditions such as pH, buffering capacity and type of acid.

The longevity of dental restoration depends on durability of the material and its properties such as hardness and wear resistance [5]. Hardness is defined as the resistance of a material to indentation [6]. Surface hardness correlates well to compressive strength, and abrasion resistance [7].

So it's valuable to study the surface roughness and the microhardness of different resin composite formulations, especially Nanofilled and BulkFilled Composites after immersion in citric acid that represent popular diet, taking into consideration the immersion time and different pH values.

Materials and Methods: One conventional composite (Filtek Z350 nanofill [NF]) and a bulk-fill composite (Filtek Bulk-fill [BF] were used in the current study.

Immersion media: Distilled water for control groups and as storage medium neutral pH (6.5-7) and Citric acid solutions prepared with pH values of 2.5 and 5.5.

Preparation of Specimens: A Split Teflon mold was used to prepare the specimens with dimensions 5.0 mm in diameter and 2.0 mm in thickness. A total of 168 specimens (5 mm × 2 mm) were fabricated 84 for each type of composite material. Then each of these groups will be further divided into 3 smaller subgroups (28 specimens for each) according to the pH value of the immersion solution either neutral (6.5-7) (distilled water), (2.5 citric acid solution) and (5.5 citric acid solution). Then each of these smaller groups will be further divided into 2 subgroups according to the time of immersion either 3 days or 7 days.

Testing procedures:

1. **Surface microhardness testing:** were completed using Digital Display Vickers Micro-hardness Tester (Model HVS-50, Laizhou Huayin Testing Instrument Co., Ltd. China), with a Vickers diamond indenter and a 20X objective lens

2. Surface Roughness testing: Specimens were photographed using USB Digital microscope with a built-in camera *(Scope Capture Digital Microscope, Guangdong, China)* connected with an IBM compatible personal computer using a fixed magnification of 120X. Images were recorded with a resolution of 1280 × 1024 pixels per image. WSXM software was used to calculate average of heights(Ra) expressed in μm, which can be assumed as a reliable indices of surface roughness [8].

SEM analysis

SEM studies were carried out on specimens of Nanofilled and Bulk Filled Composites before and after acid immersion to examine change in the surface microstructure of the composite materials.

Statistical analysis:

The mean and standard deviation values were calculated for each group in each test. Data were explored for normality using Kolmogorov-Smirnov and Shapiro-Wilk tests, data showed parametric (normal) distribution.

Independent sample t-test was used to compare between two groups in non-related samples. Paired sample t-test was used to compare between two groups in related samples. One-Way ANOVA followed by Tukey post hoc test was used to compare between more than two groups in non-related samples. Three-way ANOVA was used to test the interaction between different variables.

Results: 1- Microhardness test: Data in table (2) shows the results of three-way ANOVA analysis for the interaction of different variables. The results showed that different pH had no statistically significant effect at P-value 0.812. Also, immersion time had no statistically significant effect at P-value 0.231. Different materials had no statistically significant effect at P-value 0.963. The interaction between the three variables also had no statistically significant effect at P-value =0.050.

Table (1): Three-way ANOVA for the effect of different variables on hardness evaluation.

Source of variation	Type III sum of Squares	df	Mean Square	F - value	P-value
pH	72.824	2	36.412	.209	.812ns
Time	252.945	1	252.945	1.449	.231ns
Materials	0.380	1	0.380	0.002	.963ns
pH x Time x Materials interaction	1064.193	2	532.097	3.048	.050ns

2-Surface Roughness Test: Data in table (3) shows the results of three-way ANOVA analysis for the interaction of different variables. The results showed that different pH had no statistically significant effect at P-value 0.926. Also, time had no statistically significant effect at P-value 0.555. While different materials had a statistically significant effect at P-value 0.025. The interaction between the three variables also had no statistically significant effect at P-value =0.480.

Table (3): Three-way ANOVA for the effect of different variables on surface roughness.

Source of variation	Type III sum of Squares	df	Mean Square	F - value	P-value
pH	1.181E-06	2	5.904E-07	0.0770921	0.926ns
Time	2.675E-06	1	2.675E-06	0.3493338	0.555ns
Materials	3.944E-05	1	3.944E-05	5.1499535	0.025*
pH x Time x Materials interaction	1.13E-05	2	5.648E-06	0.7374218	0.480ns

Discussion:

In the current study, results revealed that Nanofilled composite **(NF)** exhibited higher VHN (Vicker's Hardness number) than that of the bulk filled composite (**BF)** when immersed in neutral distilled water that might be attributed to the higher inorganic filler content in **(NF)** with a less filler loading according to the manufacture's description and previous studies [9,10]. The composite resin Z350 XT has a combination of zirconia and silica nanoparticles. Furthermore, these particles form nanoclusters (0.6 to 1.4 μm). These nanoclusters act like a single filler [11]. All of these factors justify the increased VHN of **NF** than **BF** in our control groups [12,13]. After immersion in citric acid solutions it was found that as the pH value of the solution decreases (5.5 then 2.5), **BF** was more resistant to degradation than NF and exhibited increased VHN values.

Considering time of immersion, either 3 or 7 days, there were no significant differences between the two materials but results revealed that **(BF)** showed slight more resistance to degradation as immersion time increased. The lower susceptibility of (**BF)** to degradation by citric acid solutions might be attributed to the type of monomers other than matrix (BisGMA). **(BF)** is based on urethane dimethacrylate (UDMA) and also contained aromatic urethane dimethacrylate (AUDMA) and addition fragmentation monomers (AFM) for moderating stiffness of its polymer matrix. Also larger filler sizes might also explain the greater resistance to degradation and higher VHN in comparison to conventional composite **(NF)** considering time factor [14]. Regarding **(NF),** the Bis-GMA monomer is considered the most viscous and least flexible monomer due to the strong intramolecular hydrogen bonding of its hydroxyl groups (-OH) on the backbone and the presence of rigid aromatic nuclei interactions given by aromatic rings in its structure. UDMA is also a viscous monomer due to the hydrogen bond intramolecular interaction between its Amine (--NH--) and carbonyl groups (-C==O). However, the viscosity of UDMA is much lower and flexibility is higher in relation to Bis-GMA because of the weak hydrogen bond of its Amine group compared to that of hydroxyl groups [15].

Also, the presence of Amine groups in the urethane structure of UDMA monomer was responsible for the characteristic chain transfer reactions that provided an alternative path for the continuation of polymerization. These reactions result in increased mobility of radical sites on the network and consequently enhanced polymerization and monomer conversion [16]. This explained the increased reactivity and higher Degree of conversion of UDMA containing organic matrix of **(BF)** when compared to that of Bis-GMA containing organic

matrix of **(NF)** [17] that would directly improve the surface hardness of the material.

Considering the effect of pH on VHN of **NF** and **BF**, it was shown that samples that were soaked in the acidic mediums of either 2.5 or 5.5 had greater VHN values than that of the control samples soaked in distilled water. It is well known that the surface roughness against the polyester matrix film produces the smoothest surface for most tooth colored restorations. Though, this surface is polymer rich making it quite unstable. The polymer rich layer is commonly clinically removed by finishing and polishing which produce surface irregularities that increase the roughness to the polished surface of varying degrees depending on the polishing systems and materials used [18]. In the current study no polishing procedures were carried out, so it is supposed that when the samples were soaked in the acidic solutions the resin rich layer was removed exposing more fillers. This justifies the greater VHN values than the control samples soaked in distilled water.

So, it's highly recommended to perform finishing and polishing procedures following composite application to avoid resin-rich superficial layer that maybe completely eroded leaving fillers that are exposed and causes rough surface of the composite[19,20]. This in turn result in plaque retention, periodontal diseases and recurrent caries [21].

Ra value (surface roughness) is the arithmetic mean of the departures of the roughness profile from the mean line [22]. According to Hamouda [22], the roughness of all intra-oral hard surfaces should approximate a Ra value of 0.2 μm or lower to reduce bacterial retention.

Regarding surface roughness, it was shown that non-significant statistical difference was detected between **NF** and **BF.** The values have shown that **NF** exhibited higher surface roughness values than **BF** in all immersion groups either in neutral,2.5 or 5.5 pH**.** This could be justified as since nano-filled composites contain greater surface area to volume ratio of their fillers particle system, which may cause them to suffer higher surface roughness or degradation as compared to other resin based materials [23].

These changes also might be attributed to sorption of water by **(NF)** under acidic conditions leading to an increase in roughness, as it's composed predominantly of monomers that are more susceptible to hydrolysis, that is, Bis-GMA and TEGDMA. But the particles that are stripped out from the surface are very small; hence, they leave small holes that produced an insignificant increase in roughness values [24,25].

This was in accordance with **Aftab Ahmed Khan et al** [23]**,** who conducted a study that evaluated the effect of different beverages on surface topography and hardness of nano-filled composite materials and found that nano-filled composites possess higher surface area to volume ratio of their fillers particle size that might lead to higher surface roughness than other resin based dental materials.

Surface Roughness values revealed that **NF** and **BF** had increased surface roughness after immersion in acidic solutions of 2.5 and 5.5 pH values respectively. The effect of water uptake of **BF** & **NF** could promote its degradation [26] because water absorption caused hydrolysis of silane coupling agents and loss of chemical bonds between filler particles. Resin matrices could also promote dislodgement of filler particles from the outer surface, resulting in rapid increase in surface roughness and facilitating the erosion of the composite

This was in agreement with **Hamouda** [22] who reported that all restorative materials tested in his study became rougher after they had been subjected to the lower pH-cycling regimen.

Conclusions and Recommendations:

1. pH value of the solution negatively affects surface characteristics of dental composites.

2. Short immersion periods (3 or 7 days) were not sufficient to investigate the acid effect of on surface properties of dental composites so it's recommended to extend the time of immersion in further studies.

3. Lack of finishing and polishing step dramatically affected the surface characteristics of dental composites.

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