Surface roughness evaluation of nanofiller composite resin after immersion in 50% calamansi orange juice (Citrus microcarpa)

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ABSTRACT Nano-filled composite resin is a composite resin that has nano-sized fillers. This filler size gives an aesthetic appearance to the composite resin thus resembles natural teeth. However, composite resin has a disadvantage in its polymer matrix, which is easily degraded by acid solutions. Calamansi orange (Citrus microcarpa) is a solution that has acidic properties. The purpose of this research was to determine the surface roughness value of nanofiller composite resin after immersed in a 50% concentration of calamansi orange (Citrus microcarpa). Ten cylindrical specimens of nano-filled composite resin (Filtex Z350 XT 3M ESPE), with 5 mm diameter and 2 mm thickness. The specimens were immersed for 14 days with the provisions of 5 minutes in the juice of calamansi orange and 23 hours 55 minutes in distilled water. Calamansi orange juice is replaced every day. The evaluation of surface roughness of specimens before and after immersion using Atomic Force Microscopy (AFM). Furthermore, the data were analyzed using paired t-test with a significant value is p <0.05. The results showed an increase in the average value of surface roughness which was significant after exposure in the calamansi orange with 50% concentration.

KEYWORDS: Atomic Force Microscopy, Citrus microcarpa, surface roughness, nanofilled composite

INTRODUCTION

Composite resin is increasingly being used as a dental restoration material because it has a color similar to natural teeth, does not dissolve easily, and has better physical properties than other restorations. A composite resin material that has a color similar to natural teeth is nanofiller composite resin. Nanofiller composite resins have an outstanding filling component that gives them translucency properties similar to natural teeth and improves the material's mechanical strength. However, this material is easily degraded by acid solutions.

Degradation by the acid solution in the nanofiller composite resin resulted in breaking the polymer filler chain. Research by Dentiana et al. (2016), which carried out immersed of nanofiller composite resin in a 1.5 ml solution, showed that the mean concentration of Bis-GMA monomer release in acidic solution immersion (pH 3.5) was higher than alkaline solution (pH 8) and neutral solution (pH 7). As a result of the release of the Bis-GMA monomer in the immersion of the acid solution, it causes the instability of the chemical bonds of the double chain polymer matrix. The instability of this chemical bond causes the double chain of the polymer matrix to become broken and degraded, causing bumps on the filler surface. Research by Roque et al. (2015) showed that the spots on the filler make the surface of the nanofiller composite resin rougher after being exposed to acid solutions compared to those not exposed to acid solutions.

Tantanuch et al. (2014) showed an increase in surface roughness of the nanofiller composite
resin after immersion in an acid solution in patients using nanofiller composite resin restorations. One of the acidic solutions that people often consume is calamansi orange (Citrus microcarpa). Calamansi orange (Citrus microcarpa) has a sour taste from citric acid and vitamin C, which is higher than other types of orange. Calamansi orange (Citrus microcarpa) has various health benefits such as reducing levels of fatty acids and cholesterol (anti-hypercholesterolemia), as an antihypertensive and preventing multiple chronic diseases such as cancer.

Surlitah showed a decrease in the lipid profile in overweight adult women using calamansi orange (Citrus microcarpa) with a concentration of 50%. Setiawan et al. (2017) reported the level of community compliance in drinking calamansi orange (Citrus microcarpa) is a health drink at 50% concentration is higher than 100% concentration of calamansi orange juice. This is because the taste is more acidic in the concentration of 100% calamansi orange juice. The contact between the acid content in oranges and the nanofiller composite resin material will result in roughness on the surface of the nanofiller composite resin. Based on the description above, this research aimed to evaluate the surface roughness of the nanofiller composite resin after exposure to 50% Citrus microcarpa juice was carried out.

MATERIALS AND METHODS

This research is an experimental laboratory using the Pre Test - Post Test Group design which was conducted at the Laboratory of the Faculty of Dentistry, Syiah Kuala University, and the Laboratory of Materials Physics, Faculty of Mathematics and Natural Sciences (FMIPA), Syiah Kuala University. There are six specimens of nanofiller composite resin (Filtek Z350 XT, 3M ESPE, A3), in the form of a cylinder with 5 mm diameter and 2 mm thickness. The specimens were immersed for 14 days with the provisions of 5 minutes in calamansi orange juice and 23 hours 55 minutes in distilled water. Calamansi orange juice is replaced every day.

Preparation of Nanofiller Composite Resin Specimens

The specimens were made using a stainless steel mold that had been smeared with Vaseline. The center of the mold was filled with nanofiller composite resin and coated with a celluloid strip matrix. Next, the polymerization was done using a light-curing unit (LCU) with a 2 mm perpendicular irradiation distance for 20 seconds with 900 mW/cm² light intensity. After the specimens polymerized, the samples were stored in distilled water for 24 hours.

Production of 50% Calamansi Orange Juice

Calamansi oranges were sliced in half, squeezed the water, and then filtered. 50% concentration was obtained by mixing 25 ml of calamansi orange juice and 25 ml of distilled water. Calamansi orange juice and distilled water were homogenized and put into ten plastic vials with 5 ml each plastic vial.

pH Measurement

Measurement of acid degree (pH) was carried out three times, and the average was calculated. With the first calibration using a pH meter (Neutron tech pH-009-A).

Immersion Method

Specimens were immersed in calamansi orange with 50% concentration for 5 minutes. Then rinsed and put in distilled water for 23 hours 55 minutes. Immersion was carried out until all specimens were immersed, and the immersion was stored in an incubator at 37°C for 14 days. Specimens were immersed for 14 days with provisions of 5 minutes in calamansi orange juice and 23 hours 55 minutes in distilled water. Calamansi orange juice is changed every day.

Evaluation Procedure of Surface Roughness of Nanofiller Composite Resin

Evaluation of surface roughness values on six nanofiller composite resin specimens was carried out twice, namely before and after immersion in 50% calamansi orange juice. The roughness test was carried out with Atomic Force Microscopy (AFM) (NanoSurf 2 EasyScan). AFM will scan the surface of the specimen. Furthermore, any deflection of light on the tip will be reflected in the photodiode. The roughness result will be displayed on the computer screen.

Data Analysis

The results of surface roughness evaluation of nanofiller composite resin before and after immersion in calamansi orange juice at 50% concentration were analyzed and processed using a paired t-test with Statistical Product and Service Solution (SPSS) software version 24.
RESULTS

The results showed an increase in the surface roughness of nanofiller composite resin after immersion in calamansi orange juice (Citrus microcarpa) at 50% concentration, and the average pH measurement results were 3.53 (Table 1).

| No. | Specimen | Roughness (µm) (x ± SD) | P |
|-----|----------|-------------------------|---|
| 1   | Before   | 0.055 ± 0.018           |   |
| 2   | After    | 0.108 ± 0.075           | 0.002* |

*significant difference (paired t-test p<0.05)

DISCUSSION

This research showed an increase in the average surface roughness of the nanofiller composite resin before and after immersion in calamansi orange juice at 50% concentration. It indicates the presence of roughness after immersed in calamansi orange at 50% concentration. The average value of surface roughness of nanofiller composite resin before immersion in calamansi orange juice at 50% concentration was 0.055 m. At the same time, the average value of surface roughness of nanofiller composite resin after immersion in the calamansi orange juice at 50% concentration was 0.075 m. Based on the results of the paired t-test, it was found that there was a significant difference in the results of surface roughness evaluation of nanofiller composite resin before and after immersion in calamansi orange juice at 50% concentration.

The increase in average surface roughness of nanofiller composite resin after immersion in calamansi orange juice at 50% concentration was thought to be influenced by acid content in the calamansi orange juice and the physical properties of water absorption possessed by the nanofiller composite resin. The acid content of calamansi orange at 50% concentration was 3.53. It was because calamansi orange juice contains citric acid, ascorbic acid, and vitamin C. The degree of acidity indicates that the 50% concentration of calamansi orange juice was acidic (pH<7).

Acid solutions could accelerate the hydrolysis process, where acid solutions had a chemical molecular structure with excess H+ ions. H+ ions through the protonation process are bound to water molecules (H2O) become hydroxonium ions (H3O+). This protonation process occurred in the silane coupling agent bond and matrix ester bond (Bis-GMA, TEGDMA, and UDMA). The ester bond is reacted with H3O+ ions by transferring protons so that free carboxylic acid groups are formed, reducing the acidity in the nanofillers composite resin matrix. The acidic conditions in the matrix can degrade and damage the matrix structure, causing the release of monomers and accelerating debonding or release between matrix with filler in nanofiller composite resin. Research by Dentiana et al. (2016) the release of Bis-GMA monomer in nanofiller composite resins was greater in specimens immersed in acid solution (pH 3.5) than in neutral solution (pH 7).

Release or degradation of filler from nanofiller composite resin is also affected by water absorption. Water absorption occurs in the polymer matrix in the form of TEGDMA, where the matrix is susceptible to absorption and solubility while in contact with acid solutions. TEGDMA has the greatest hydrophilic property, namely 69.51µg/mm3. This hydrophilic nature will provide water access into the polymer matrix and binds to the monomer, causing the polymer matrix to debond and hydrolytic degradation with the nanofiller composite resin filler material. As a result of this, water absorption can cause the surface roughness of the composite resin to increase.

The figure below shows the topography of the surface roughness of the nanofiller composite resin (a) before and (b) after immersion in the calamansi orange juice at 50% concentration. It can be seen that light colors (mountains) indicate high surfaces, and dark colors (valleys) indicate deep characters from the topography. While the brownish-red color indicates the surface between peaks and valleys. The more profound the feeling read by AFM, the higher the surface roughness value, and the shallower the surface read by AFM, the slightly lower roughness value. This refers to the method...
of calculating the surface roughness value. Surface roughness is calculated by the vertical deviation of the entire surface from the expected value. If the deviation value is large, the surface is rough. Conversely, if the deviation value is small, then the surface is slightly rough.25

![Figure 1. Microscopic view of topography using Atomic Force Microscopy (AFM) (a) before and (b) after immersion in calamansi orange juice at 50% concentration.](image)

The deep surface (valley) shows part of the specimen surface eroded by the acid solution. Seen in Fig 1(a) and Fig 1(b) on specimen number 7 shows many valleys based on topographical images after immersion in 50% concentration of calamansi orange juice compared to topographical images before immersion in 50% calamansi orange juice. It indicates an increase in the roughness seen from the topographic image after immersion in Citrus microcarpa juice at 50% concentration compared to before immersion.

The specimen's topography shows a dark color (valley), presumably in the form of pores formed on the sample's surface due to exposure to calamansi orange, which contains acid. Soman et al. (2019) found that acetic acid, citric acid, and malic acid in apple cider can produce surface roughness in nanofiller composite resins.20 Supported by Bajwa’s research, et al. (2014), there was an increase in the roughness of nanofiller composite resins in acidic media due to the absorption of water from the nanofiller composite resin followed by hydrolysis process on the silane coupling agent bond, which is increased potential for debonding or released filler particles. The release of filler particles can produce holes marked by an increase in the surface roughness of the nanofiller composite resin.26

Based on the results of the research, the average surface roughness value of the nanofiller composite resin, it can be concluded that consuming 50% concentration of the Citrus microcarpa is still recommended because the value shown is not significant and the roughness value shown is still below the standard roughness value that can be accepted by the oral cavity, which is less than 0.2 µm.

**CONCLUSION**

Based on the results of the research, it can be concluded that there is an increase in the average roughness of the nanofiller composite resin after immersion in a cycle of 5 minutes in calamansi orange juice at 50% concentration with a 3.53 pH average within 14 days.
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