The Effect of Different Surface Treatments on Tensile Bond Strength of Acrylic Denture Teeth and Light-cured Resin composite

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Abstract

Acrylic denture teeth often exhibit wear or fracture which may lead to a loss of mastication efficiency and aesthetically unpleasant. Light-cured resin composite can be used to repair the acrylic denture teeth. However, the research published on the tensile bond strength between acrylic denture teeth and resin composite is very limited. The purpose of this study is to investigate the effect of different chemical surface treatments on tensile bond strength of acrylic denture teeth and resin composite. Sixty maxillary lateral incisors acrylic denture teeth were embedded with labial surface downwards and ground with silicon carbide paper. The teeth were divided into six groups (n=10): no surface treatment (control), methyl methacrylate treatment (MMA) for 180 seconds, methyl formate-methyl acetate mixture (MF-MA*)* solution at a ratio of 25:75 (v/v) treatment for 15 seconds, composite bonding agents, MMA (180 seconds) with the application of bonding agent, and MF-MA (15 seconds) with a bonding agent. Tensile strength was measured using a universal testing machine at a crosshead speed of 0.5mm/min. The data were analyzed using one-way ANOVA and post hoc Dunnett T3 test (α=0.05). The surface treatments significantly affected the tensile bond strength between acrylic denture teeth and resin composite ($p<0.05$). The bond strengths of the control group, MMA treated, and MF-MA treated group was not significantly different ($p>0.05$). The bond strength of the MF-MA with the bonding agent group was significantly higher than the other group ($p<0.05$). This study suggests the application of MF-MA solution for 15 s followed by a composite bonding agent before repair procedure can increase the tensile bond strength between acrylic denture teeth and resin composite.

Keywords: *Acrylic denture teeth, Resin composite, Methyl acetate, Methyl formate, Tensile bond strength*

1. Introduction

Acrylic resin teeth are often preferred to fabricate removable complete and partial dentures due to their structure that similar to denture base material structure, so they can chemically bond to the denture base (Anusavice, Phillips, Shen, & Rawls, 2013). Moreover, acrylic denture teeth are easy to adjust, it does not wear the natural opposing teeth, does not cause a clicking sound when chewing, and they can provide a satisfying aesthetic result. Nonetheless, acrylic denture teeth debonding, fracture and wear can occur over time and require the repair procedure. It has been reported that fracture and debonding damages were the common failure of the acrylic denture and were accounted for 26% (Vallittu, Lassila, & Lappalainen, 1993) to 33% (Darbar, Huggett, & Harrison, 1994) of all denture repairs. In order to repair the broken acrylic denture teeth, the in-office procedure may be proposed because it is less time consuming, less cost, aesthetically pleasing, and it is not required that the patient is without prosthesis.

Light-cured resin composite can be used for prosthodontic purposes in various applications including the replacement of lost or broken denture teeth, the correction of acrylic resin denture teeth wear, and the modification labial surfaces of denture teeth to harmonize esthetic of the prosthetic teeth with the adjacent natural teeth (Lagouvardos & Polyzois, 2003; Papazoglou & Vasilas, 1999; Vergani, Machado, Giampaolo, & Pavarina, 2000). In an attempt to use resin composites for repairing the acrylic resin teeth, the chemical bond may not occur since acrylic denture teeth are highly polymerized. Consequently, the surface treatments of acrylic denture teeth are required to provide micromechanical retention and ensure the longevity of the repaired denture.

Previously, many chemical solutions have been used to treat acrylic denture teeth surfaces such as chloroform, methylene chloride, 4-methacryloxyethyl trimellitate anhydride (4-META), and methyl methacrylate (MMA). MMA is the most frequently used chemical agent for treating the surface of acrylic denture teeth. MMA has been proved that it can promote the bond strength of repaired acrylic denture base, relined acrylic denture base, and the bond strength between acrylic denture teeth and acrylic resin. Moreover, there were studies about the effect of using MMA as a chemical surface treatment to the bond strength between acrylic denture teeth and resin composite. It had been reported that surface treatment with MMA enhanced the bond strength between acrylic denture teeth and resin composite (Papazoglou & Vasilas, 1999), especially when combined with the use of a bonding agent(Chatterjee, Gupta, & Banerjee, 2011; Muhsin, 2017; Papazoglou & Vasilas, 1999; Vergani et al, 2000; Yanikoglu, Duymus, & Bayindir, 2002). Since chloroform and methylene chloride were identified as being carcinogenic, Asmussen and Peutzfeldt (2000) investigated the effect of several chemical solvents on the softening efficiency in an attempt to use as denture base softening agent substitutes for those carcinogenic solutions. They found that low molecular weight methyl esters such as methyl formate (MF) and methyl acetate (MA) were as effective as methylene chloride (Asmussen & Peutzfeldt, 2000).

Vallittu, Lassila, and Lappalaina (1994) suggested that in order to achieve the effective primed surface and diminish the adhesive failure, the best wetting time for repair heat-polymerized acrylic resin using MMA as surface treatment is 180 seconds (Vallittu, Lassila, & Lappalainen, 1994). Whereas, a study revealed that a 15-seconds MF-MA treated acrylic denture teeth had significantly higher micro-tensile bond strength between the denture teeth and heat-cured denture base material, compared with no surface treatment teeth (Penpattanakul, Arunpraditkul, & Wiwatwarapan, 2018). Another study found that increased wetting time of MF-MA (15, 30, 60, and 180 seconds) did not increase the tensile bond strength of relined denture base resin (Tanasamanchoke & Wiwatwarapan, 2015). A similar result was found in the study of the tensile bond strength between auto-polymerized acrylic resin and acrylic denture teeth treated with MF-MA using various application time. The tensile bond strength of the 15-second MF-MA treated group was significantly higher than the no-treatment group but was not significantly different compared with the MF-MA 30 seconds, 60 seconds, 120 seconds, 180 seconds, and the 180-second MMA treated group (Thongrakard & Wiwatwarrapan, 2016). The effect of different concentrations of MF-MA mixture solution had been studied, most of the reports showed that the 25:75 %v/v concentration was the best concentration to promote the bond strength of the acrylic denture base (Leklerssiriwong & Wiwatwarapan, 2012; Osathananda & Wiwatwarrapan, 2016; Penpattanakul & Wiwatwarapan, 2012; Thunyakitpisal, Thunyakitpisal, & Wiwatwarapan, 2011).

The MF, MA, and MF-MA mixture also increased the bond strength of repaired acrylic denture base, acrylic denture base relined with rebasing material, and the bond strength between acrylic denture teeth and auto-polymerized acrylic resin. Thus, it is hypothesized that the mixture of MF-MA could improve the tensile bond strength between acrylic denture teeth and resin composite.

Past studies indicated that using MF-MA solutions that is a non-toxic surface treatment agent can enhance the bond strength of acrylic resin material with the advantage of less time-consuming. However, there is no study of the effect of using MF-MA solution to the bond strength between acrylic denture teeth and resin composite. Moreover, past studies about the effect of different surface treatment on the bond strength between acrylic denture teeth and resin composite were focus only on the shear bond strength. The purpose of this study was to investigate and compare the effect of the surface treatment with MMA, MF-MA, and composite bonding agent on the tensile bond strength between acrylic denture teeth and resin composite.

2. Objectives

To evaluate and compare the effect of different chemical surface treatments on tensile bond strength of acrylic denture teeth and resin composite.

3. Materials and Methods

The acrylic denture teeth, resin composite, and chemical agents used in this study are shown in Table 1. Sixty lateral incisors of acrylic resin denture teeth (Yamahachi New Ace, Japan) were embedded with labial surface down in auto-polymerized potting resin and were packed in 20-mm diameter polyethylene pipes. The labial surfaces of denture teeth were polished, to a depth of proximately 1-1.5mm, with 600-grit silicon carbide paper in a polishing machine (Nano2000, PACE Technologies, St. Tucson, AZ, USA). The specimens were divided into groups of 10 specimens each and were ultrasonically cleaned in distilled water for 10 minutes. Chemical surface treatments were applied to the groups as follows:

Group I: No surface treatment was performed (negative control group). A clear polypropylene film with a 3-mm diameter hole was placed over the surface.

Group II: MMA (Unifast Trad, GC Dental Product Corp, Aichi, Japan) was applied for 180 seconds (by brush every three seconds). The excess MMA was then air-blown away. A clear polypropylene film with a 3-mm diameter hole was placed over the treated surface.

Group III: MF-MA mixture solution at a ratio of 25:75 by volume was applied for 15 seconds (by brush every three seconds). The excess liquid was then blown away. A clear polypropylene film with a 3-mm diameter hole was placed over the treated surface.

Group IV: A clear polypropylene film with a 3-mm diameter hole was placed over the cleaned surface. Two layers of composite bonding agent (Adper Single Bond 2 Adhesive, 3M ESPE, St. Paul, MN, USA) were applied for 15 seconds, the air thinned, and light-cured for 20 seconds.

Group V: MMA monomer was applied for 180 seconds (by brush every three seconds). After the excess MMA was blown away, a clear polypropylene film with a 3-mm diameter hole was placed over the treated surface. Two layers of composite bonding agent were applied for 15 seconds, the air thinned, and light-cured for 20 seconds.

Group VI: MF-MA solution was applied for 15 seconds (by brush every three seconds). After the excess liquid was blown away, a clear polypropylene film with a 3-mm diameter hole was placed over the treated surface. Two layers of composite bonding agent were applied for 15 seconds, the air thinned, and light-cured for 20 seconds.

All specimens were covered with a 5-mm diameter acrylic resin ring that placed centrally over the hole of polypropylene film. Resin composite (Filtek Z350XT, 3M ESPE, St. Paul, MN, USA) was then placed in the ring hole with 2 mm in height and light-cured for 40 seconds. After the photopolymerization process, the specimens were kept in distilled water at 37˚C for 24 hours before testing.

After the storage period, each specimen was left air dry and then was attached with acrylic resin rods using cyanoacrylate glue (Super Glue, Alteco Chemical PTE Ltd., Japan) (Fig 1). The tensile strengths were measured by a universal testing machine (Shimadzu, EZ-S 500N mode, Japan) with a 500N load cell at a crosshead speed of 0.5mm/min. Specimens were connected to the tensile testing machine by using the acrylic resin rods and secured in a vertical position (Fig 2). The tensile bond strength values (MPa) were calculated by dividing the failure force by the bond surface area.

The data were statistically analyzed using SPSS software version 22.0 (IBM corporation, New York, NY, USA). The mean and standard deviation (SD) for the tensile bond strength of each group was calculated and statistically analyzed using one-way analysis of variance (ANOVA) and the post hoc Dunnett T3 test at the 95% confidence level.

To determine the mode of fracture failure, all fracture surfaces were examined using a stereomicroscope (Olympus SZH10) at 35X magnification. In the present study, modes of failure were categorized as adhesive at the interface, mixed primarily adhesive, mixed primarily cohesive in bonding layer, mixed primarily cohesive in resin composite layer, and mixed primarily cohesive in denture teeth. There was no pure cohesive failure in this study.

Table 1 Materials used in this study

Material	Product name	Composition	Manufacturer
Denture teeth	Yamahachi New Ace	Poly(methyl methacrylate)	Yamahachi Dental Mfg.Co.,Aichi Pref., Japan
Resin composite	Filtek Z350 XT	Bis-GMA, Bis-EMA, UDMA, TEGDMA, zirconia and silica particles (5-20nm nonagglomerated)	3M ESPE, St. Paul, MN, USA
Chemical agents	Unifast Trad (liquid)	Methyl methacrylate	GC Dental product corp., Aichi., Japan
	Methyl formate	Methyl formate	Merck Schuchardt OHG, Germany
	Methyl acetate	Methyl acetate	Merck KGaA, USA
	Adper Single bond 2	Ethyl alcohol, Bis-GMA, silane treated silica, HEMA, hydroxy-1,3-dimethacryloxypropane, UDMA	3M ESPE, St. Paul, MN, USA

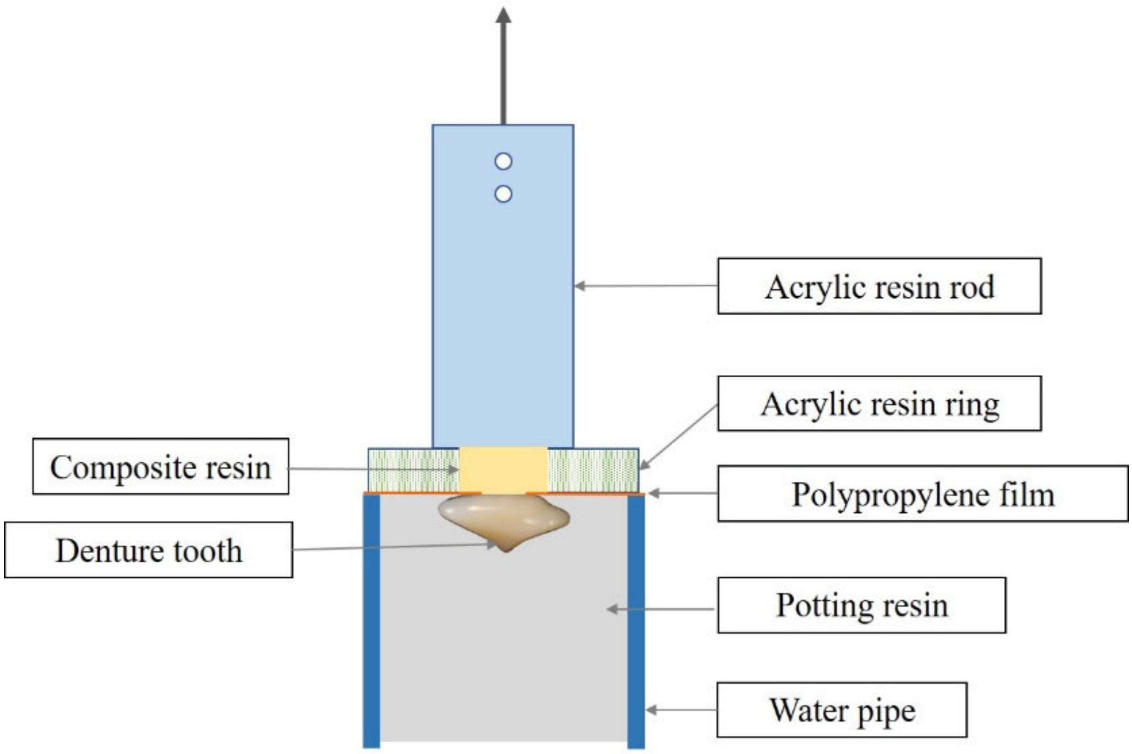

Figure 1 The schematic presentation of a specimen prepared for tensile bond strength test

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Figure 2 Sample placed for tensile bond strength test in a universal testing machine

4. Results and Discussion

The data were tested for normality using the Kolmogorov-Smirnov test and found that all data were normally distributed in all groups. The mean tensile bond strengths and standard deviation of each group are presented in Table2. The results demonstrated that the tensile bond strengths of the negative control group (group I), MMA group (group II), and MF-MA group (group III) were not significantly different from each other (p>0.05). The tensile bond strengths of the tested groups, that composite bonding agent was used as a part of surface treatment method (group IV, V, VI), were significantly higher than the groups that did not involve the use of composite bonding agent and were significantly different from each other. The tensile bond strengths of the samples treated with MMA and bonding agent (group V) along with the samples treated with MF-MA and bonding agent (group VI) were significantly higher than those treated with a bonding agent alone (group IV) (p<0.001). Whereas, The MF-MA with bonding agent group (group VI) had the significantly highest tensile bond strengths (p<0.001).

The mode of failure is presented in figure 3. All specimens in group I (negative control), group II (MMA treated), and group III (MF-MA treated) were 100% adhesive failure. The failures of specimens in group IV (bonding agent treated) were predominantly mixed primarily adhesive failure (60%) due to the majority of fracture was at the interface. The pure adhesive failure was not found in group V (MMA with bonding agent) and group VI (MF-MA with a bonding agent). Most of the specimens using MMA or MF-MA with a bonding agent as surface treatment resulted in mixed primarily cohesive failure in a bonding agent (80% and 60%, respectively) which the fracture surface showed a larger area coated with the bonding agent.

Table 2 The means tensile bond strengths (MPa) with standard deviations of each surface treatment

Group	Surface treatment	Mean tensile bond strength ± SD (MPa)
I	Control	0.43 ± 0.07^{a}
II	MMA	0.40 ± 0.08^{a}
III	MF-MA	0.43 ± 0.11^{a}
IV	Bonding agent	6.75 ± 1.62^{b}
V	MMA with a bonding agent	10.78 ± 2.28^{c}
VI	MF-MA with a bonding agent	15.52 ± 2.40^{d}

[*]The same superscript letter indicates no significant difference (p>0.05)

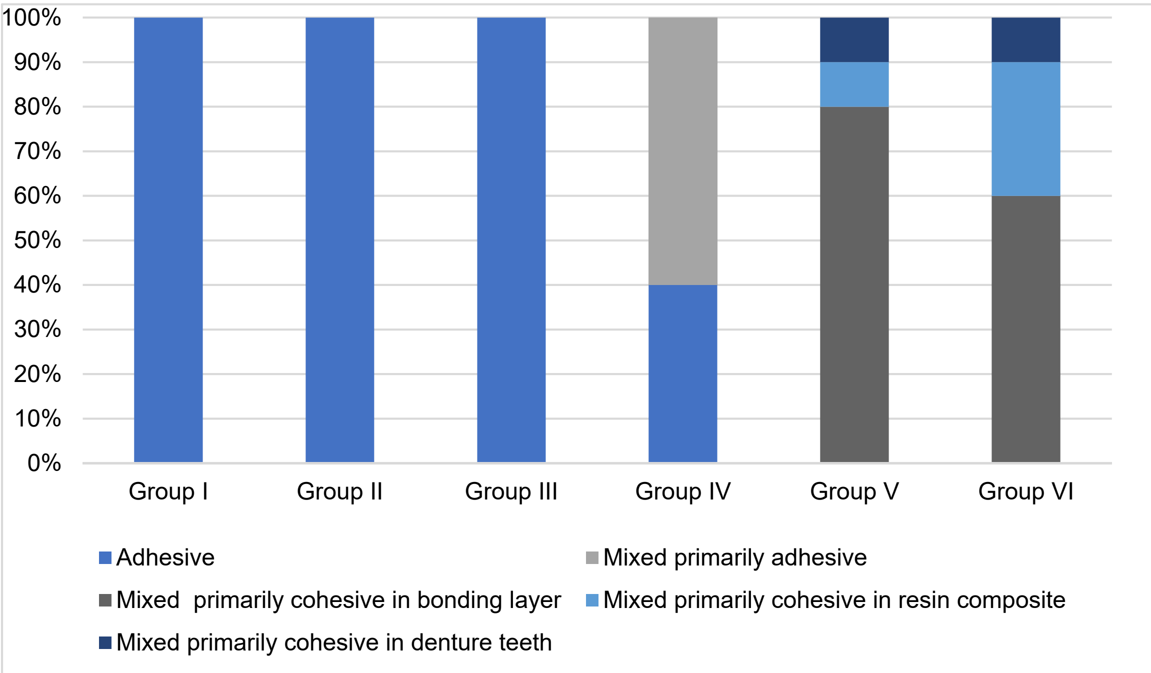

Figure 3 Failure mode in percentage (%)

Using light-cured resin composite, as the in-office procedure for repair broken or fracture acrylic denture teeth has many advantages such as less time consuming, less cost, aesthetically satisfying, and sufficiently strong. However, the bond strength between acrylic denture teeth and resin composites will directly affect the success and longevity of the repaired denture. Acrylic resin and resin composite have similar reactive methacrylate molecules (Cook, Beech, & Tyas, 1985). Therefore, the polymerization process of MMA and Bis-GMA in the resin composite matrix will follow a similar pattern of activation and cross-linking. Papazoglou and Vasilas (1999) suggested that some chemical bonding between acrylic resin and resin composite may occur if the acrylic resin teeth surface provided cross-links for bonding to composite. When acrylic resin denture teeth were bonded to resin composite, there is probably a combination of chemical and mechanical bonding (Papazoglou & Vasilas, 1999). Nonetheless, the degree of conversion of acrylic denture teeth is relatively high because of the manufacturing process. By using heat curing or cross-linking methods, this can result in rarely occurred chemical bonding.

To improve the mechanical retention between the acrylic denture teeth and auto-polymerized acrylic resin, surface treatment with chemical solvents such as MMA, methylene chloride, chloroform, ethyl acetate, and 4-META can be used. Although methylene chloride and chloroform have been used, they have been identified as noxious compounds and carcinogenic potential. Asmussen and Peutzfeldt (2000) introduced the low molecular weight methyl esters including methyl formate and methyl acetate that were as effectively softening the polymethacrylate denture base as methylene chloride but less toxic (Asmussen & Peutzfeldt, 2000). MMA monomer is the most frequently used as the chemical surface treatment agent. Since MMA has a similar chemical structure to the denture base material, so MMA can act as an adhesion promoter effectively. MMA can enhance the bond strength of acrylic denture teeth by the swelling phenomenon. When the solvent contacts the denture teeth, it will dissolve the surface and causing swelling of the surface layer. Consequently, the denture surface becomes more irregular and the monomer of the auto-polymerized acrylic material can penetrate the irregularities causing micromechanical retention (Vallittu, Ruyter, & Nat, 1997). Per contra, the surface treatment with MMA requires 180 seconds of wetting time to effectively prime the surface and ultimately reducing adhesive failures (Vallittu, Lassila, & Lappalainen, 1994)

Polymer dissolving and swelling occur when the solubility parameters and polarities of the polymer and solvent are close to each other. The solubility parameter of acrylic denture teeth material (PMMA) is 18.3 $MPa^{1/2}$, whereas MMA, MF, and MA have solubility parameters approximately 18.0, 20.9, and 19.6 $MPa^{1/2}$, respectively (Grulke, 1999). Thongrakard and Wiwatwarrapan (2016) reported that the bond strengths between acrylic denture teeth and auto-polymerized acrylic resin when using MF-MA mixture solution as a surface treatment for 15, 30, 60, 120, and 180 seconds were not significantly different compared with the using of MMA for 180 seconds. They also suggested the application of MF-MA solution for 15 seconds as an alternative chemical surface treatment for repairing a denture base and rebonding acrylic denture teeth with auto-polymerized acrylic resin (Thongrakard & Wiwatwarrapan, 2016). The centers of disease control and prevention (CDC) reported that MMA and MF have similarly 8-hour workday exposure limits which are approximately 100 ppm (NIOSH recommended exposure limit and OSHA permissible exposure limit)) but MA has higher exposure limit which is 200 ppm (NIOSH recommended exposure limit and OSHA permissible exposure limit)(Centers for Disease Control and Prevention). According to the New Jersey department of health and senior services, the odor threshold of MMA is 0.049 ppm whereas the odor threshold of MF and MA is 2,000 ppm and 180 ppm, respectively (New Jersey Department of Health and Senior Services). Thus, the benefits of using MF-MA solution as a surface treatment method are not only less application time but also less tissue irritation and less toxic.

A Past study of the effect of MF-MA surface treatment on the strength of repaired acrylic denture base revealed that the application of MF-MA mixture solution at different concentrations including 75:25, 50:50 and 25:75 %v/v resulted in similar flexural strength of the repaired denture base, however, the 25:75 %v/v concentration showed significantly higher strength when compared to MMA treatment but the others had the comparable strength to the MMA treated group (Thunyakitpisal, Thunyakitpisal, & Wiwatwarapan, 2011). Another study reported that the tensile bond strengths of acrylic denture teeth and denture base using various concentrations of MF-MA mixture solution (25:75, 40:60, and 55:45 % v/v) treatment had significantly higher bond strength than the untreated surface. The MF-MA mixture solution at a concentration of 25:75 %v/v demonstrated higher bond strength than the others although it was not significantly different (Penpattanakul, Arunpraditkul, & Wiwatwarapan, 2018). A similar result was presented by Osathananda and Wiwatwarrapan (2016), the shear bond strength of relined acrylic denture base using the 25:75 %v/v MF-MA surface treatment was the highest, followed by the using of 15:85 and 35:65 %v/v concentration (Osathananda & Wiwatwarrapan, 2016). Several studies reported the success of using 25:75%v/v MF-MA solution as the surface treatment method to increase the tensile bond strength of relined acrylic denture base (Tanasamanchoke & Wiwatwarapan, 2015), the tensile bond strength between the denture base and acrylic soft lining resin (Leklerssiriwong & Wiwatwarapan, 2012), the tensile bond strength between acrylic denture teeth and the denture base (Thongrakard & Wiwatwarrapan, 2016), and the flexural strength of relined denture base (Puangpetch & Wiwatwarapan, 2019).

According to Papazoglou and Vasilas (1999), the shear bond strength between acrylic denture teeth and resin composite can be improved by the application of MMA for 3 minutes (Papazoglou & Vasilas, 1999). Despite the fact that, in this study, the tensile bond strengths of group II (MMA-treated) and group III (MF-MA-treated) were not significantly different from group I (no treatment) and the mode of failure was completely adhesive. The results indicated that using MMA or MF-MA alone to swell the acrylic denture teeth surface did not provide sufficient micromechanical retention when repaired with resin composites. This could be influenced by the poor wettability property of the high viscosity resin composite, so the material cannot penetrate the microstructure occurring from the swelling phenomenon.

Composite bonding agent had been used to enhance the bond strength of resin composite restoration in operative dentistry. One of the important particularities of bonding agents is its low viscosity due to the presence of their solvents and diluted monomers, which enables them to penetrate deeper. Using a bonding agent to treat acrylic denture teeth before repair with resin composite, it could improve the surface wettability and promote the penetration and infiltration of the resin composite. Studies showed that the bonding agent significantly improved the shear bond strength of composite-repaired-denture teeth (Lagouvardos & Polyzois, 2003; Renne, 2010; Souza et al, 2011; Vergani, Giampaolo, & Cucci, 1997). Furthermore, several studies found that using the composite bonding agent with prior treatment of MMA for 3 minutes increased the shear bond strength between acrylic denture teeth and resin composite (Chatterjee, Gupta, & Banerjee, 2011; Muhsin, 2017; Papazoglou & Vasilas, 1999; Vergani et al, 2000; Yanikoglu, Duymus, & Bayindir, 2002). The bond was caused by the diffusion and polymerization of MMA across the acrylic tooth-resin composite interface to form interpenetrating polymer networks (Chatterjee, Gupta, & Banerjee, 2011). This study showed a similar result, the groups that using bonding agent as the surface treatment (group IV) had significantly higher tensile bond strength than the non-using bonding agent groups (group I, II, III) and most of the failure modes (60%) were mixed primarily adhesive failure. These findings indicate that a bonding agent should be used as part of surface treatment when repair acrylic denture teeth with resin composite. Using Adper Single Bond 2 not only improve the surface wettability but may also promote the micromechanical retention. The ethyl alcohol component in Adper Single bond 2 can swell and dissolve the denture teeth surface then evaporate which resulted in the surface microstructure allowing other components containing carbon-carbon double bonds (C=C) molecules penetrate deeper and polymerized to the resin composite monomer.

When using the combination of chemical solvent (MMA, MF-MA) and bonding agent (group V and VI), the tensile bond strengths were significantly higher than those using the bonding agent alone (group IV). Whereas group VI that samples were treated with MF-MA and bonding agent showed the significantly highest tensile bond strength. The mode of failures of group V and VI was also better than those of group IV since 100% mixed primarily cohesive failure was observed for group V and group VI. 80% of specimens using MMA and a bonding agent and 60% of specimens using MF-MA and a bonding agent as a surface treatment showed mixed primarily cohesive failure in the bonding layer with one sample of group V and two samples of group VI were mixed primarily cohesive failure in resin composite due to the majority of fracture were occurred in the resin composite, whereas 10% of MF-MA with a bonding agent treated surface (group VI) showed mixed primarily cohesive failure in acrylic denture teeth. These results could be explained by the swelling phenomenon. MMA and MF-MA dissolved and swelled acrylic denture teeth surface leading to microirregularity, so when bonding agent was applied, resin composite can be infiltrated into the microstructure and formed the micromechanical retention. Furthermore, the solubility parameters of MMA, MF, and MA are closer to the solubility parameter of PMMA than the solubility parameter of ethyl alcohol (26.0 $MPa^{1/2}$)(Grulke, 1999), therefore MMA and MF-MA can swell and dissolve the denture teeth surface better than Adper single bond 2. Thanyakitpisal et al (2011) investigated the characteristic of the treated acrylic denture base using a scanning electron microscope and found that MMA created shallow pits and small crest patterns, while the mixture of MF-MA created 3D honeycomb appearance (Thunyakitpisal, Thunyakitpisal, & Wiwatwarapan, 2011). These 3D pores of MF-MA-treated surface should allow greater penetration of the repaired material and hence increased surface contact and mechanical interlocking which resulted in higher bond strength.

The present study is an in vitro study that may be limited in predicting the success of a technique in clinical use. Several factors in the oral cavity that may influence the bond degradation such as saliva which contains numerous molecules with a high affinity for adsorption to the denture surface and may interfere in the bonded area. The complex denture-oral environment conditions, as well as the long periods of water storage or thermocycling, should be investigated in the future. Although this experimental method does not imitate the intraoral condition, it does provide an effective means comparing the influence of different surface treatments on the tensile bond strength between acrylic denture teeth and resin composite. The result of the present study also showed the superiority of a bonding agent in promoting adhesion between acrylic denture teeth and resin composite.

5. Conclusion

Within the limitation of the present study, the following conclusions can be drawn:

1. Using MMA or MF-MA mixture solution alone was not improve the tensile bond strength between the acrylic denture teeth and resin composite when compared to the negative control group.

2. The application of the bonding agent had a significant influence on the tensile bond strength between the acrylic denture teeth and resin composite.

3. The application of the bonding agent with prior treatment of MF-MA mixture solution (25:75 %v/v) for 15 seconds on the acrylic denture teeth resulted in the highest tensile bond strength with resin composite.

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