Effect of HTPB-TAC primer on the bond strength of a resin cement to zirconia

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Abstract

Objective: The purpose of this study was to evaluate the effect of Hydroxyl-terminated polybutadiene grafting with Trimellitic anhydride chloride (HTPB-TAC) primer on microtensile bond strength of a resin cement to zirconia ceramics.

Materials and methods: Fully sintered zirconia blocks were primed with either HTPB-TAC primer or Alloy primer® then bonded to resin composite block using a resin cement. The blocks bonded with resin cement without primer served as a negative control. Each cemented block was cut into 8 microbars (1x1mm). Pool of 48 microbars for each condition was prepared. Half of microbars in each condition were investigated with a microtensile bond strength test. The others were subjected to thermocycling for 10,000 cycles (5 to 55°C) before testing. Data were analyzed with two-way ANOVA and Games-Howell test at $\alpha = 0.05$.

Results: The microtensile bond strength values were significantly affected by primer and thermocycling without their interaction. The microtensile bond strength of HTPB-TAC primer and Alloy primer showed significantly higher bond strength compared to the non-primed specimen ($p<0.05$) while those of HTPB-TAC primer had no statistical difference from Alloy primer ($p>0.05$). The bond strength of all primers including non-primer was drastically decreased after thermocycling.

Conclusion: HTPB-TAC primer could promote the bond strength of resin cement to zirconia.

Key words: microtensile bond strength, all-ceramic, zirconia primer, zirconia, resin cement, HTPB-TAC

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Introduction

Several all-ceramic systems have been developed in dentistry to meet the expectations of patients for high aesthetic and long-lasting restorations. The zirconia core ceramics became more popular especially in multiple fix partial dentures[1]. Although bonding to silica based ceramics gave a predictable result, the physical properties and composition of zirconia based ceramics differ from silica based ceramics and require additional adhesive techniques to enhance their bonding[1,2].The conventional etching technique to ceramics using hydrofluoric acid follows by the application of silane coupling is effective method for silica based ceramics. The chemical bond between the inorganic phase of ceramic and the organic phase of the resin was enhanced by these agents. However, zirconia ceramics cannot be etched by hydrofluoric acid since ceramics do not contain glassy matrix in their structure[2].

Recent studies[2,3]revealed that clinical failures of all-ceramic crown have initiated from the cementation or internal surfaces of the crown. This matter can be overcome by the application of reliable chemical bonding of the luting agent to the crown. It has been shown that the use of 10-Methacryloyloxydecyl dihydrogen phosphate (10-MDP) containing luting agents enhanced the bond strengths to zirconia ceramics. It was found that 10-MDP monomer may bond chemically to zirconium oxide layer on the zirconia surfaces[4-7].

An experimental primer, Hydroxyl-terminated polybutadienegrafting with Trimellitic anhydride chloride (HTPB-TAC), which has a carboxylic acid terminal as an active functional group, was recently developed[8]. It was found that the carboxylic acid derivative monomer could improve the bond strength of resin cements to silica based ceramics and also zirconia ceramics[9]. Therefore, the purpose of this study was to evaluate the effect of the HTPB-TAC primer on microtensile bond strength of a resin cement to zirconia ceramic.

Materials and methods

Hydroxyl-terminated polybutadiene (HTPB, Ricon120®, Sartomer, PA, USA) was grafted with Trimellitic anhydride chloride (TAC, SM Chemical Supplied Co.Ltd., Bangkok, Thailand), at HTPB:TAC ratio = 1:2 by mol. The reaction was done at 65°C for 24 hr. The un-reacted TAC was precipitated by methanol. The HTPB-TAC product was filtrated and dried at 50°C for 2 hr. Two percent by weight of HTPB-TAC in acetone was used as an experimental primer in this study. The chemical structure of HTPB-TAC was showed in Figure 1. The Alloy primer (Kuraray Medical Inc., Tokyo, Japan) which contain 10-MDP as an active monomer, was used as positive control primer.

Fully sintered yttrium-oxide–partially-stabilized zirconia blocks (ZENO®, Weiland, Pforzheim, Germany), size 8 x 5.5 x 4 mm were prepared. The blocks were sandblasted on the bonded surface with 250 μm and 50 μ m aluminum oxide particles respectively. Resin composite blocks (Z350®, 3M-ESPE, St.Paul, MN, USA), size 8 x 5.5 x 4 mm were also prepared. The composite blocks were polished on the bonded surface using 600 grit silicon carbide papers under water cooling. Both zirconia and composite blocks were cleaned in an ultrasonic cleaner for 10 minutes followed by vapor-streamed cleaner for 5 seconds.

The bonded surface of zirconia was primed either with the Alloy primer or experimental primer by 3 rounds painting with paintbrush then allowed its reaction and dried for 1 min. Non-primer specimens were used as negative control. RelyX U-100 cement (3M-ESPE, St.Paul, MN, USA) was used as a luting cement. The cement pastes were hand

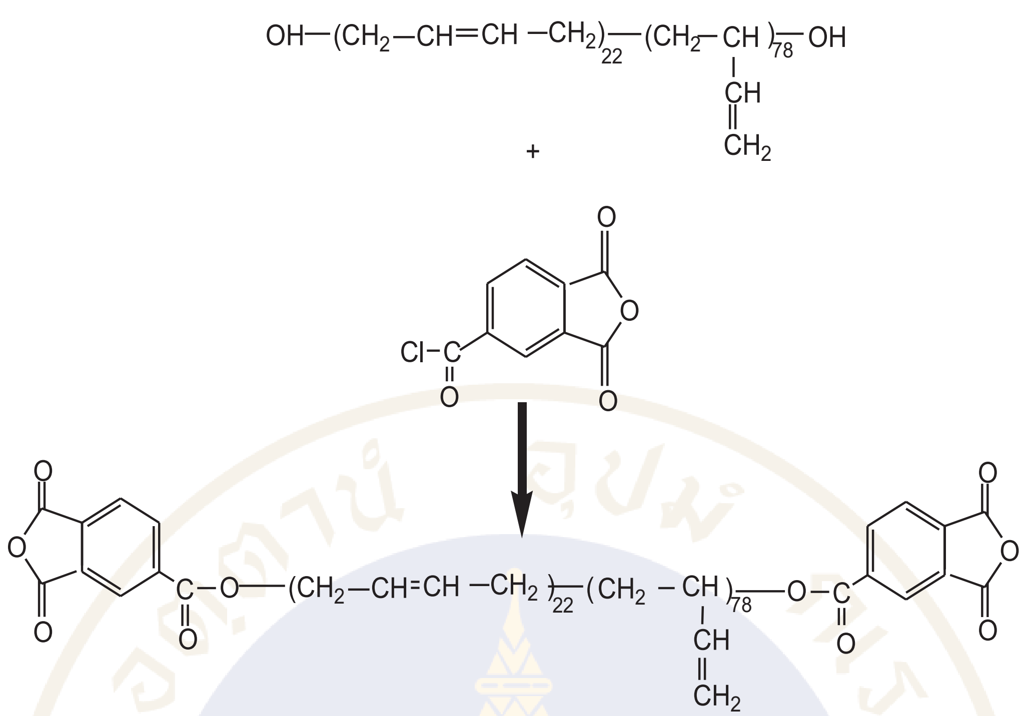

Figure 1 Chemical formula of HTPB-TAC primer

mixed according to the manufacturer's instruction for 20 s, and then applied on the composite block. The composite block was then bonded to the treated zirconia surface under 20 N load, and then light polymerized for 20 s on each side. 6 specimen blocks were prepared for each condition.

The bonded blocks were glued with cyanoacrylate to a metallic base of low-speed diamond precision cutting machine (Accutom-50, Struers, Ballerup, Denmark). Each block was sectioned 1 mm interval both vertically and horizontally to prepare one square millimeter microbars with a 0.4 mm-thick diamond coated cutting disc under water cooling at a speed of 400 rpm. Eight precise microbars in the central of the block were obtained from each block. The schematic illustration of the prepared specimen and cutting was showed in Figure 2. The pooled 48 microbars for each condition were randomly assigned into two testing conditions, Thermocycling and non-thermocycling equally. For thermocycling, the specimens were subjected to thermocycling for 10,000 cycles at 5 and 55^{o}C with 30s dwelling time.

The precise dimension of the interfacial bonded area of each sample was measured using a digital caliper. The specimens in both conditions were then subjected to microtensile bond strength test using a microtensile tester (Bisco Inc, Schaumburg, IL, USA) at a crosshead speed of 1 mm/min until fracture.

The microtensile bond strength in megapascal (MPa) was calculated using load at fracture in newton (N) divided by interfacial bonded area of the specimen (mm^2). The microtensile bond strength data were statistical analyzed by two-way ANOVA and then post hoc multiple comparisons by Game-Howell test at 95% confidence level.

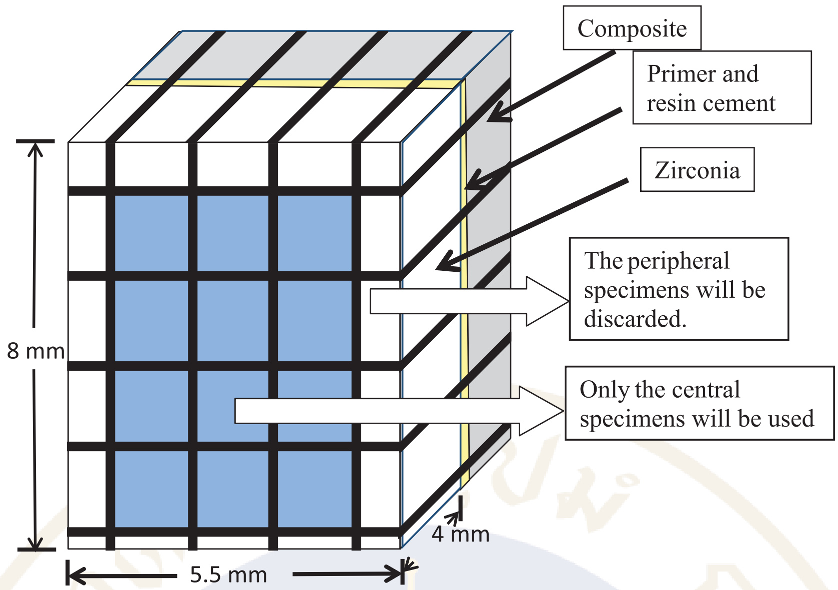

Figure 2 The schematic illustration of the prepared specimen and specimen cutting

The fracture surfaces of all specimens were examined under an optical light microscope (Nikon Eclipse E400, Nikon Corp, Tokyo, Japan) at 40x magnification. Three types of failure were defined[7].

1. Cohesive failure: fracture occurred completely either within resin cement, composite or zirconia

2. Adhesive failure: fracture occurred clearly at the resin cement-composite or cement-zirconia interface

3. Mixed failure: fracture occurs in both cohesive and adhesive failure.

Results

The means and standard deviations of microtensile bond strength and type of failure of all specimens were shown in Table 1 and Figure 3.

Two-way ANOVA clearly showed that the microtensile bond strength between zirconia and composite was influenced by both main factors, type of primer and thermocycling process ($p< 0.05$) without their interaction ($p>0.05$).

Thermocycling decreased the bond strength of all specimens regardless to prime or non-prime condition ($p<0.05$). Figure 3 obviously revealed the effect of thermocycling process for each experimental group.

HTPB-TAC primer could promote the bond strength of resin cement to zirconia significantly compared to non-primer group ($P<0.05$). However, there was no significant difference between the bond strength of group using Alloy primer and HTPB-TAC primer ($p>0.05$).

For mode of failure analyses as shown in Figure 4, the non-primed group before thermocycling showed mixed failure with some composite remained on the zirconia surface. However, after thermocycling all specimens in the non-primed group revealed adhesive failures with the surface of zirconia ceramic were completely exposed after fracture. While the other groups, demonstrated mixed failures regardless to thermocycling. There were no specimens presented the cohesive failure in zirconia ceramic or within resin cement.

Table 1 Microtensile bond strength (MPa) of resin cement to zirconia and type of failure

		Microtensile bond strength (MPa±SD)	Type of failure		
			Adhesive	Cohesive	Mixed
Non-primer	Non-themocycling	10.2 ± 3.8[b]	0	0	24 (100%)
	Thermocycling	3.4 ± 2.2[c]	24 (100%)	0	0
Alloy primer	Non-themocycling	19.9 ± 4.4[a]	0	0	24 (100%)
	Thermocycling	11.2 ± 1.9[b]	0	0	24 (100%)
HTPB-TAC primer	Non-themocycling	17.3 ± 2.5[a]	0	0	24 (100%)
	Thermocycling	8.8 ± 2.6[b]	0	0	24 (100%)

Bond strength values with same superscript letters indicate no statistically significant difference

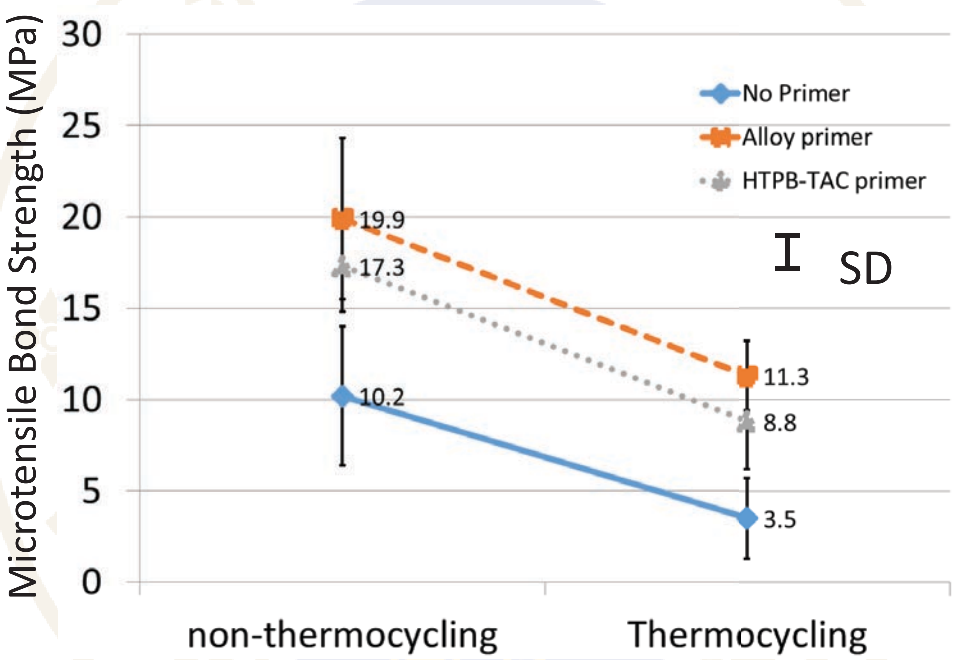

Figure 3 Microtensile Bond Strength of resin cement to zirconia and thermocycling effect

Discussion

The HTPB-TAC primer was firstly developed as an alloy primer to enhance the bonding of dental alloy to tooth structure[8]. The structure of HTPB-TAC primer contain trimellitic anhydride terminal in both end of polybutadiene core structure. Trimellitic anhydride functional group was successfully proved that it can hydrolyze to carboxylic acid functional groups and developed a chemical bond to alloy as in 4-META[8]. However, in modern ceramic as zirconia, only 10-MDP showed high initial bond strength to zirconia[9-12] among commercial available adhesive primers,. However, the bond strength was quite low for clinical application and unreliable to thermocycling or water storage[13,14]. Therefore, 10-MDP were selected to use as a positive control in this study. The bonding of 10-MDP to zirconia was purposed as same as its bonding to alloy by forming a crosslink with zirconium oxide layer[5-7].It was interested to study whether the HTPB-TAC primer which has double carboxylic functional group in its structure can bond well to oxide layer of zirconia ceramic.

According to this study, the highest bond strength values were obtained with Alloy primer (19.9 ± 4.4 MPa) followed by HTPB-TAC primer (17.3 ± 2.5 MPa). However, there were no statistical significant difference among both primers ($p>0.05$). Both primers gave a higher bond strength than non-primer application (10.2 ± 3.8 MPa) ($p<0.05$). The mode of failure of all specimens before thermocycling showed mixed failure. This means that HTPB-TAC primer could promote the bond between zirconia and resin composite comparable to commercial available primer.

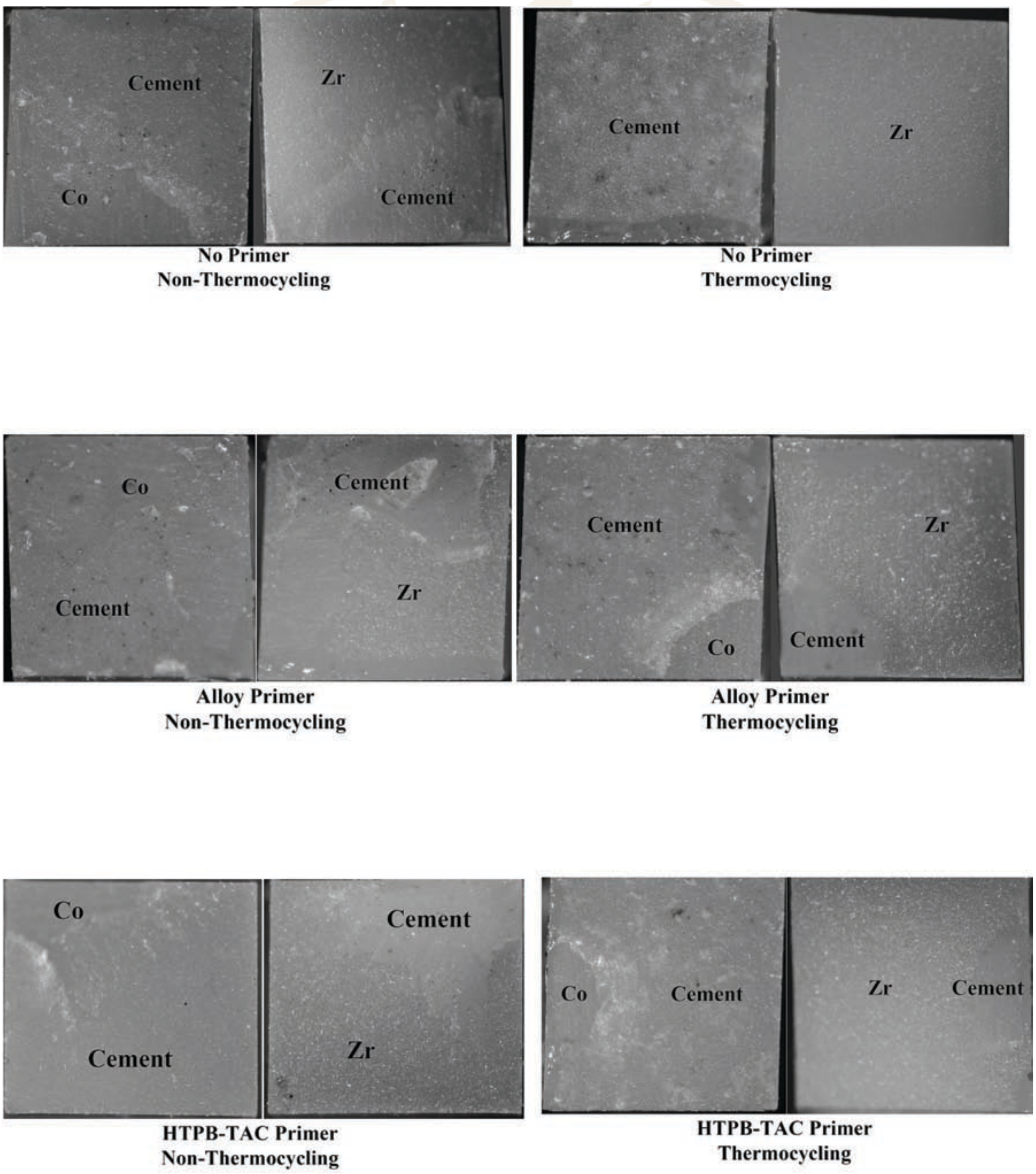

Figure 4 The representative microscope pictures of fracture surfaces on composite and zirconia site of the specimen, with different primers and thermocycling.

However, the bond strength of all primers including non-primer was drastically decreased after thermocycling. The bond strength was decreased equally regardless of primer (figure 3). Several previous studies revealed that thermocycling process significantly reduced bond strength of resin cement to zirconia[7,15,16]. The adhesive ability of the acidic primer to zirconia are depend on the presence of a zirconium-oxide passive coating on the ceramic surface[5,7]. Although before thermocycling, the bond strength of resin cement to zirconia could establish without the fact that a true chemical bond with zirconia was established or whether it relies on micro-retention provided by particle abrasion[17]. However, the mode of failure of non-primer after thermocycling showed the different mode of failure compare to others. The mode of failure of non-primer group changed from mixed mode to adhesive mode after thermocycling, while those of both primer groups still showed mixed mode. Some studies found that micro-retention bonding and some primer bond linkage are not durable for hydrolytic effects in oral condition[7,18]. This imply that the application of primers was gave higher and chemical reliable bond strength to zirconia than non-primer application.

In conclusion, this study showed that HTPB-TAC primer has a potential of enhancing bonding zirconia to resin composite. The thermocycling process played a significant role in degradation of the bond. To use HTPB-TAC primer intra-orally, future studies of the stability and biocompatibility of HTPB-TAC primer are required.

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References

1. Raigrodski AJ. Contemporary materials and technologies for all-ceramic fixed partial dentures: a review of the literature. *J Prosthet Dent* 2004; 92: 557-62.
2. Borges GA, Sophr AM, de Goes MF, Sobrinho LC, Chan DC. Effect of etching and airborne particle abrasion on the microstructure of different dental ceramics. *J Prosthet Dent* 2003; 89: 479-88.
3. Bottino MA, Valandro LF, Scotti R, Buso L. Effect of surface treatments on the resin bond to zirconium-based ceramic. *Int J Prosthodont* 2005; 18: 60-5.
4. Yoshida K, Tsuo Y, Atsuta M. Bonding of dual-cured resin cement to zirconia ceramic using phosphate acid ester monomer and zirconate coupler. *J Biomed Mater Res B Appl Biomater* 2006; 77: 28-33.
5. Kern M, Wegner SM. Bonding to zirconia ceramic: adhesion methods and their durability. *Dent Mater* 1998; 14: 64-71.
6. de Souza GM, Silva NR, Paulillo LA, De Goes MF, Rekow ED, Thompson VP. Bond strength to high-crystalline content zirconia after different surface treatments. *J Biomed Mater Res B Appl Biomater* 2010; 93: 318-23.
7. Oyagüe RC, Monticelli F, Toledano M, Osorio E, Ferrari M, Osorio R. Effect of water aging on microtensile bond strength of dual-cured resin cements to pre-treated sintered zirconium-oxide ceramics. *Dent Mater* 2009; 25: 392-9.
8. Sanohkan S. *Development of universal adhesive primer for maxillofacial application* (Doctor of Philosophy). Dental Biomaterials, Mahidol University. Bangkok: Mahidol University; 2011.
9. Komine F, Kobayashi K, Saito A, Fushiki R, Koizumi H, Matsumura H. Shear bond strength between an indirect composite veneering material and zirconia ceramics after thermocycling. *J Oral Sci* 2009; 51: 629-34.
10. Urapepon S, Wootthisarnwatthana K. Effect of noble metal primers on the microtensile bond strength of resin cement to zirconia. *Mahidol Dent J* 2011; 31: 65-72.

11. Kim MJ, Kim YK, Kim KH, Kwon TY. Shear bond strengths of various luting cements to zirconia ceramic: surface chemical aspects. ***J Dent*** 2011; 39: 795-803.
12. Dias de Souza GM, Thompson VP, Braga RR. Effect of metal primers on microtensile bond strength between zirconia and resin cements. ***J Prosthet Dent*** 2011; 105: 296-303.
13. de Sá Barbosa WF, Aguiar TR, Francescantonio MD, Cavalcanti AN, de Oliveira MT, Giannini M. Effect of water storage on bond strength of self-adhesive resin cements to zirconium oxide ceramic. ***J Adhes Dent*** 2013; 15: 145-50.
14. de Souza G, Hennig D, Aggarwal A, Tam LE. The use of MDP-based materials for bonding to zirconia. ***J Prosthet Dent*** 2014; 112: 895-902.
15. Lüthy H, Loeffel O, Hammerle CH. Effect of thermocycling on bond strength of luting cements to zirconia ceramic. ***Dent Mater*** 2006; 22: 195-200.
16. D'Amario M, Campidoglio M, Morresi AL, Luciani L, Marchetti E, Baldi M. Effect of thermocycling on the bond strength between dual-cured resin cements and zirconium-oxide ceramics. ***J Oral Sci*** 2010; 52: 425-30.
17. Aboushelib MN, Matinlinna JP, Salameh Z, Ounsi H. Innovations in bonding to zirconia-based materials: Part I. ***Dent Mater*** 2008; 24: 1268-72.
18. Valandro LF, Della Bona A, Antonio Bottino M, Neisser MP.The effect of ceramic surface treatment on bonding to densely sintered alumina ceramic. ***J Prosthet Dent*** 2005; 93: 253-9.