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Improvement of enamel bond strengths for conventional and resin-modified glass ionomers: acid-etching vs. conditioning*

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Abstract: Objective: This study deals with the effect of phosphoric acid etching and conditioning on enamel micro-tensile bond strengths (μTBSs) of conventional and resin-modified glass ionomer cements (GICs/RMGICs). Methods: Forty-eight bovine incisors were prepared into rectangular blocks. Highly-polished labial enamel surfaces were either acid-etched, conditioned with liquids of cements, or not further treated (control). Subsequently, two matching pre-treated enamel surfaces were cemented together with one of four cements [two GICs: Fuji I (GC), Ketac Cem Easymix (3M ESPE); two RMGICs: Fuji Plus (GC), RelyX Luting (3M ESPE)] in preparation for μTBS tests. Pre-treated enamel surfaces and cement-enamel interfaces were analyzed by scanning electron microscopy (SEM). Results: Phosphoric acid etching significantly increased the enamel μTBS of GICs/RMGICs. Conditioning with the liquids of the cements produced significantly weaker or equivalent enamel μTBS compared to the control. Regardless of etching, RMGICs yielded stronger enamel μTBS than GICs. A visible hybrid layer was found at certain enamel-cement interfaces of the etched enamels. Conclusions: Phosphoric acid etching significantly increased the enamel μTBSs of GICs/RMGICs. Phosphoric acid etching should be recommended to etch the enamel margins before the cementation of the prostheses such as inlays and onlays, using GICs/RMGICs to improve the bond strengths. RMGICs provided stronger enamel bond strength than GICs and conditioning did not increase enamel bond strength.

Key words: Glass ionomer cements, Surface treatments, Bovine enamels, Scanning electron microscopy (SEM), Micro-tensile bond strengths

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1 Introduction

Glass ionomer cements (GICs) are composed of calcium fluoroaluminosilicate glasses and an aqueous solution of polyelectrolyte (Wilson and Kent, 1972; Sidhu, 2011; Moshaverinia *et al*., 2012). The latter is a homo- or co-polymer of unsaturated carboxylic acids (Wilson and Kent, 1972; Moshaverinia *et al*., 2012).

Compared with other restorative materials, GICs exhibit several clinical advantages, including physicochemical bonding to tooth structures (Glasspoole *et al*., 2002), fluoride release over a long period (Kent *et al*., 1979; Khouw-Liu *et al*., 1999), and good biocompatibility (Tanumiharja *et al*., 2000; Xie *et al*., 2000). Moreover, GICs are considered smart materials, self-adhering to tooth hard tissues without any surface pretreatment, due to the ionic bond formation between the hydroxyapatite (HAp) of tooth hard tissues and carboxyl groups of polyalkenoic acid (Wilson and Kent, 1972; Yoshida *et al*., 2000).

However, GICs have some disadvantages, such as low early strength and moisture sensitivity (Fajen *et al*., 1990; Wiltshire, 1994; Khouw-Liu *et al*., 1999; Itoh *et al*., 1999; Xie *et al*., 2000; Chitnis *et al*., 2006).

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In order to improve the chemical and physical properties of GICs, resins were incorporated into GICs. As a result, resin-modified glass ionomer cements (RMGICs) were introduced to the dental profession in 1988, which contain fluoroaluminosilicate glasses, polyacrylic acid, as well as a methacrylate monomer, such as hydroxyethyl methacrylate (HEMA) (Antonucci *et al*., 1988; Nicholson and Czarnecka, 2008).

Surface pretreatments are of great importance for dental adhesion and cementation. Enamel bond strengths depend on the etching agents, acid concentration, and etching time (Bishara *et al*., 2000b; Movahhed *et al*., 2005; Espinosa *et al*., 2010). Phosphoric acid etching on enamel surfaces has been widely used as a standard bonding procedure using etch-and-rinse adhesives (Buonocore, 1955; de Munck *et al*., 2005; van Meerbeek *et al*., 2010; Pashley *et al*., 2011). More recently, selective enamel etching with phosphoric acid has been highly recommended to improve the performance of the mild self-etch adhesives (van Landuyt *et al*., 2006; Erickson *et al*., 2009; van Meerbeek *et al*., 2011). However, the use of selective enamel etching or phosphoric acid etching for GICs/RMGICs has not been adequately explored, probably because these materials can adhere to the tooth hard tissue surfaces by chemical mechanisms (Wilson and Kent, 1972; Yoshida *et al*., 2000; 2001; Fukuda *et al*., 2003).

A review of the dental literature reveals many controversial reports about the effect of acid-etching and conditioning on the enamel bond strengths of GICs/RMGICs (Powis *et al*., 1982; Silverman *et al*., 1995; Attin *et al*., 1996; Cacciafesta *et al*., 1998; 1999; 2003; Bishara *et al*., 2000b; Valente *et al*., 2002; Coutinho *et al*., 2006). Several investigators demonstrated that GICs/RMGICs bonded to the enamel effectively without a need to etch or condition the teeth before bonding procedures (Silverman *et al*., 1995; Cacciafesta *et al*., 1998; 1999). On the contrary, some laboratory studies revealed the improvements in the enamel bond strengths of GICs/RMGICs after enamel surface pretreatments with various solutions (Powis *et al*., 1982; Cortes *et al*., 1993; Attin *et al*., 1996; Bishara *et al*., 2000a; 2000b; Glasspoole *et al*., 2002; Valente *et al*., 2002; Cacciafesta *et al*., 2003; Coutinho *et al*., 2006).

In general, most of the previous studies focused on the surface treatments of RMGICs (Cortes *et al*., 1993; Silverman *et al*., 1995; Attin *et al*., 1996; Cacciafesta *et al*., 1998; 1999; 2003; Bishara *et al*., 2000a; 2000b; Valente *et al*., 2002; Coutinho *et al*., 2006), while few studies dealt with the pretreatments of the enamel surfaces for GICs (Powis *et al*., 1982; Glasspoole *et al*., 2002). Until recently, the effect of acid-etching or conditioning on the enamel bond strengths of GICs/RMGICs has not been investigated in detail. The null hypothesis tested in this study was that phosphoric acid etching and conditioning with the liquids of the cements would not significantly increase the enamel bond strengths of GICs/RMGICs.

Therefore, the aim of this study was: (1) to investigate the effect of phosphoric acid etching and conditioning with the liquids of the cements on the enamel micro-tensile bond strengths (μTBS) of two GICs and two RMGICs, and (2) to analyze the micromorphologies of the enamel-cement interfaces and the enamel surfaces by scanning electron microscopy (SEM) whether the polished enamel surfaces were etched, conditioned, or not further treated.

2 Materials and methods

2.1 Specimen preparations

This research protocol was carried out in accordance with the international Ethical Guidelines and Declaration of Helsinki and approved by the Ethics Committee of Zhejiang University School of Stomatology. Fifty-four non-carious bovine mandibular incisors were stored in 0.1% (w/v) thymol solution at 4 °C, and used for this study within three months after extraction. They were prepared into rectangular blocks. After all labial enamel surfaces were serially wet-ground with 300-, 600-, 1200-, and 2500-grit SiC abrasive paper for 30 s, ending with 4000-grit SiC paper for 1 min, 48 incisors were randomly assigned into four groups according to four cements [two GICs: Fuji I (GC, Japan), Ketac Cem Easymix (3M ESPE, Germany); two RMGICs: Fuji Plus (GC, Japan), RelyX Luting (3M ESPE, Germany)]. The polished enamel surfaces of all the specimens were either etched with 37% (w/v) phosphoric acid (ETCH-37, Bisco Inc., Schaumburg, IL, USA) for 15 s, conditioned with the liquids of the cements for 15 s, or not further treated (control).

Subsequently, they were water-sprayed for 30 s, and gently air-dried, before two matching pretreated enamel surfaces were luted together with one of the four cements under finger pressure for 5 min. The specimens were covered by Vaseline at the enamel-cement interfaces and stored in tap water. The materials used in the study are summarized in Table 1.

2.2 μTBS measurement

After all the specimens had been stored in tap water at room temperature for 24 h and the 24 specimens resulting from 48 incisors had been luted together, every pair of incisors were perpendicularly sectioned through the enamel-cement interfaces using a low-speed saw (Isomet 1000, Buehler, Lake Bluff, IL, USA) under continuous water cooling. The specimens were prepared into multiple beams about 9 mm long with a rectangular cross-sectional area of approximately 1 mm². The μTBS tests were performed with a micro-tensile tester (Bisco Inc., USA) at a crosshead speed of 1 mm/min until fracture. The μTBS was calculated in MPa.

2.3 Mode of failure

The failure mode of all the debonded specimens was determined under a light microscopy (Nikon Eclipse 80i, Tokyo, Japan) at 100-fold magnifications. The failure mode was categorized into three types as following: Type 1, no cements remained on the fractured surfaces, revealing a smooth enamel surface; Type 2, a few cements remained on the fractured surfaces; and Type 3, most of the fractured surfaces were covered with cement residues.

2.4 SEM

Two 1-mm thick pieces from each subgroup were obtained during the specimen sectioning for μTBS tests. The sectioned surfaces were immersed in 0.1 mol/L HCl for 10–30 s to expose the enamel-cement interfaces for SEM observations.

The enamel surfaces of another six bovine incisors were treated as per the above-mentioned for SEM observations. In addition, two debonded specimens from each subgroup were randomly selected after μTBS tests for SEM observations.

After all the specimens were dehydrated in ascending concentrations of ethanol and gold sputter-coated, the cement-enamel interfaces, fractured surfaces, and the pre-treated enamel surfaces were analyzed by SEM (ZEISS ULTRA 55, Germany).

2.5 Statistics

Statistical analysis was performed with statistical software (SPSS software, Version 17.0, SPSS Inc., Chicago, IL, USA). The data were analyzed with 3×4 factorial design analysis of variance (ANOVA). Post-hoc least significant difference (LSD) multiple comparisons were used to analyze the statistical differences in μTBS data among the four groups with different surface treatments.

Table 1 Materials used in this study

Material	P/L	Nature	Component
Etch-37 Bisco Inc., Schaumburg, IL, USA LOT: 0900005544			37% phosphoric acid
Ketac Cem Easymix 3M ESPE AG, Seefeld, Germany LOT: 376688 (P), 371540 (L)	3.8/1	GICs	Powder: glass powder, polycarboxylic acid, pigments; Liquid: water, tartaric acid, conservation agents
Fuji I GC, Tokyo, Japan LOT: 0812021 (P), 0811261 (L)	1.8/1	GICs	Powder: fluoroaluminosilicate glass, polyacrylic acid; Liquid: polyacrylic acid, water, polycarboxylic acid
Fuji Plus GC, Tokyo, Japan LOT: 0901211 (P), 0901221 (L)	2/1	RMGICs	Powder: fluoroaluminosilicate glass; Liquid: copolymer of acrylic and maleic acids, HEMA, tartaric acid, water, chemical initiators
RelyX Luting 3M ESPE AG, Seefeld, Germany LOT: N119359 (P), N119360 (L)	1.6/1	RMGICs	Powder: fluoroaluminosilicate glass; Liquid: polyalkenoic acid and HEMA, water, chemical initiators

P: powder; L: liquid; GIC: glass ionomer cement; RMGIC: resin-modified glass ionomer cement; HEMA: hydroxyethyl methacrylate

3 Results

3.1 μTBS

The μTBS data are summarized in Table 2. Compared with the control, phosphoric acid etching significantly increased the enamel μTBSs of GICs/RMGICs (P<0.01). Conditioning with the liquids of the cements produced significantly weaker enamel μTBS (Ketac Cem Easymix and Fuji Plus) (P<0.05) than or equivalent enamel μTBS (Fuji I and RelyX Luting) (P>0.05) as the control. Regardless of etching, RMGICs (RelyX Luting, Fuji Plus) yielded stronger enamel μTBS than GICs (Fuji I, Ketac Cem Easymix) (P<0.01). In addition, Fuji Plus yielded the strongest enamel μTBS among the four GICs on highly-polished enamel surfaces (control) (P<0.01).

3.2 Failure analysis

The failure analysis data are graphically presented in Fig. 1. Type 1 failure revealed a smooth surface on the fractured surfaces, occasionally occurring in this study (Fig. 2a). Type 2 failure revealed some cement residues on fractured surfaces on small amounts of fractured specimens (Fig. 2b). Type 3 failure revealed that the fractured surface was mainly covered with cement residues on the majority of fractured specimens (Fig. 2c).

3.3 SEM observation

The polished enamel surface revealed a very smooth smear layer with several scratches (Fig. 3a). The enamel surfaces etched with phosphoric acid or conditioned with the liquids of cements such as Fuji I and Ketac Cem Easymix showed a typical honeycomb etching pattern (Figs. 3b–3d). The enamel conditioned with the liquids of cements such as RelyX Luting and Fuji Plus exhibited the preferential dissolution of the interprismatic matrixes without the complete removal of the polishing scratches (Figs. 3e and 3f).

An obvious, continuous, and uniform hybrid layer was found at the enamel-cement interfaces of the etched enamel when Fuji I, RelyX Luting, and Fuji Plus were used (Figs. 4b, 6b, and 7b), but not

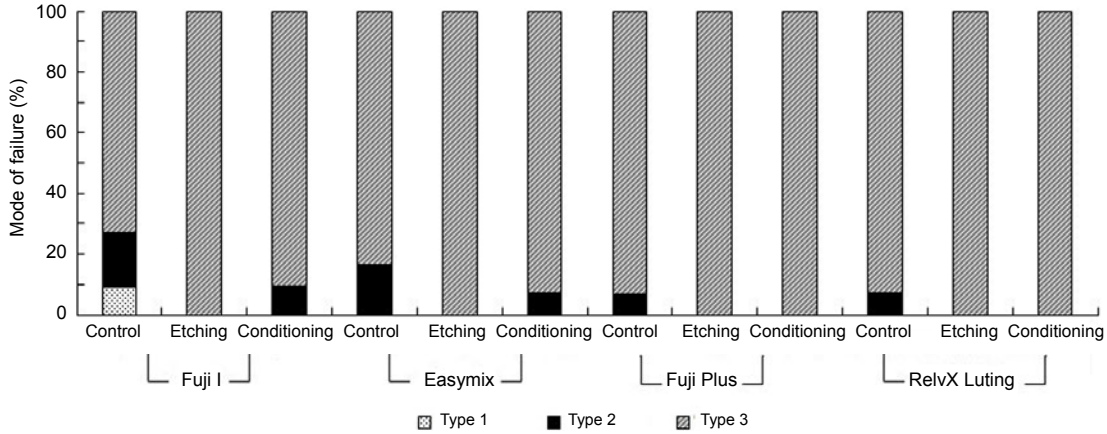


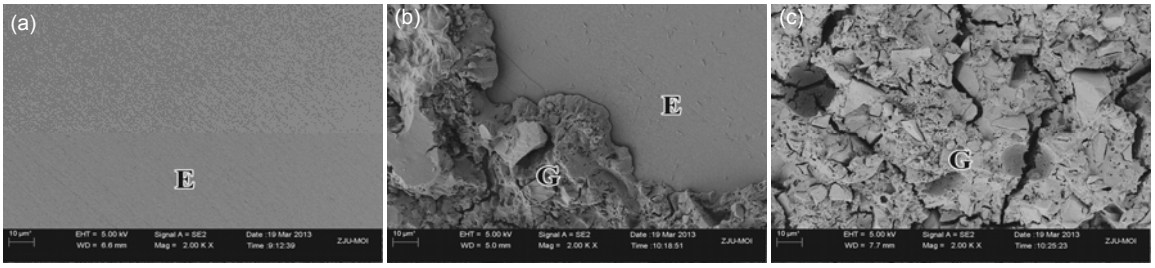
Fig. 1 Failure analysis of GICs/RMGICs after different treatments

Type 1 failure revealed a smooth surface on a fractured surface. Type 2 failure revealed some cement residues on some fractured surfaces, mainly occurring on the highly-polished enamel surfaces (control group) or on the enamel surface conditioned with liquids of GICs. Type 3 failure revealed cohesive failure of cements that fractured surfaces were mainly covered with cement residues on the majority of fractured specimens

Table 2 Mean micro-tensile bond strengths (μTBSs) to the enamels

Surface treatment	μTBS (MPa)			
	Fuji I	Easymix	Fuji Plus	RelyX Luting
Polishing	4.75±2.22 (11)^{**a}	8.27±2.08 (12)$^{\#\#a}$	22.32±3.65 (15)$^{\blacklozenge\blacklozenge b}$	15.45±5.70 (14)$^{\blacktriangle\blacktriangle g}$
Etching	11.32±3.34 (13)^{***c}	12.09±4.79 (20)$^{\#\#\#c}$	29.15±4.00 (10)$^{\blacklozenge\blacklozenge\blacklozenge d}$	27.79±5.47 (10)$^{\blacktriangle\blacktriangle\blacktriangle d}$
Conditioning	6.40±2.81 (21)^{**e}	5.29±3.52 (14)$^{\#e}$	17.02±7.00 (13)$^{\blacklozenge f}$	17.52±4.67(13)$^{\blacktriangle\blacktriangle f}$

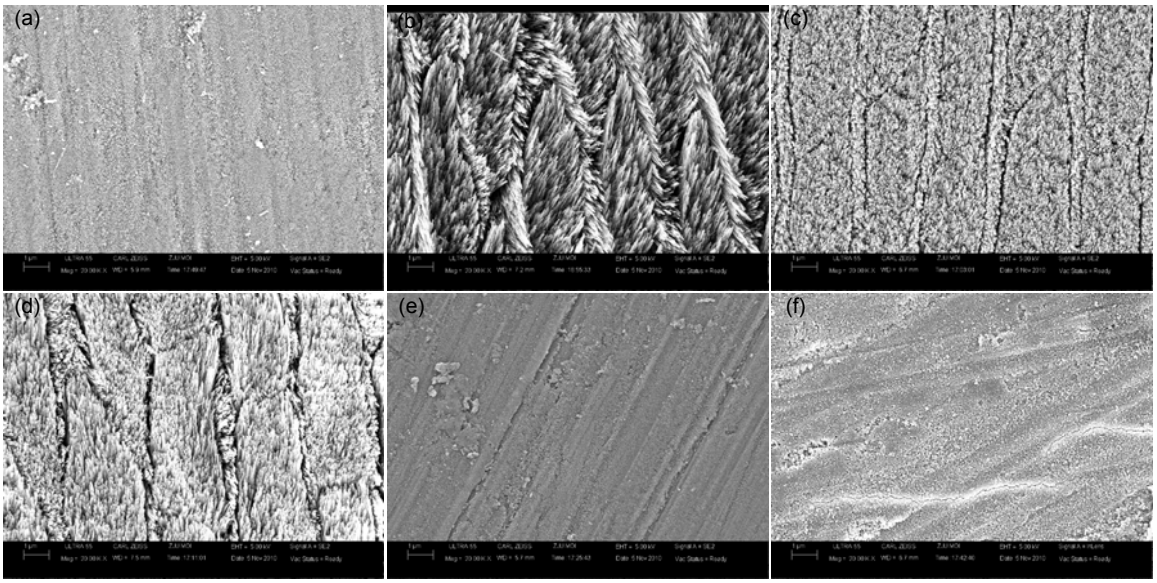
Data are expressed as mean±SD (n), analyzed by 3×4 factorial design ANOVA followed by post-hoc LSD multiple comparisons; n is the number of the specimens for μTBS tests. The different superscript letters in each column or the different superscript symbols in each row indicate significant differences (P<0.05)



Magnification=2 000-fold, bar=10 μm

Fig. 2 SEM fractography of the different fractured surfaces

SEM fractography revealed smooth surface on the fractured surface of one control specimen luted with Fuji I (a), some cements residues on fractured surfaces of a control specimen luted with RelyX Luting (b), and amount of cement residues on the fractured surface of an etched specimen luted with RelyX Luting (c). G: resin-modified glass ionomer cements (RelyX Luting); E: enamel



Magnification=20 000-fold, bar=1 μm

Fig. 3 Micromorphology of the enamel surfaces after various treatments

(a) The highly-polished enamel surface presented a smooth smear layer with some scratches; (b) The acid-etched enamel surface revealed a typical etching pattern; (c, d) The enamel surfaces conditioned with the liquids of Fuji I (c) or Ketac Cem Easymix (d) showed the complete removal of the smear layer and preferential dissolution of interprismatic matrixes; (e) The enamel surfaces conditioned with the liquids of Fuji Plus looked like the polished enamel surface; (f) Conditioning with the liquid of RelyX Luting produced the shallow pittings scattered on the smear layer

detected when Ketac Cem Easymix was used (Fig. 5b). Furthermore, typical resin tags penetrating into the partially decalcified enamel substrate with network structures were found at the cement-enamel interfaces of the etched enamels, forming the thick hybrid layers between the cements and the etched enamels (Figs. 4b, 6b, and 7b). In contrary, the hybrid layers at the enamel-cement interfaces were thin or almost invisible when the enamel surfaces were merely highly polished (control) (Figs. 4a, 5a, 6a, and 7a) or they were further conditioned with the liquids of the cements (Figs. 4c, 5c, 6c, and 7c).

4 Discussion

In this study, bovine incisors were used as a substitute for human teeth to evaluate the enamel bond strengths of GICs/RMGICs because bovine enamel possesses similar chemical compositions, physical properties, and microstructure as human enamel (Feagin *et al*., 1969; Putt *et al*., 1980; Yassen *et al*., 2011; Wang *et al*., 2012). Furthermore, bovine and human enamel possess the same bonding/cementing behavior with dental adhesives and cements (Nakamichi *et al*., 1983; Jiang *et al*., 2010; Yassen *et al*., 2011).

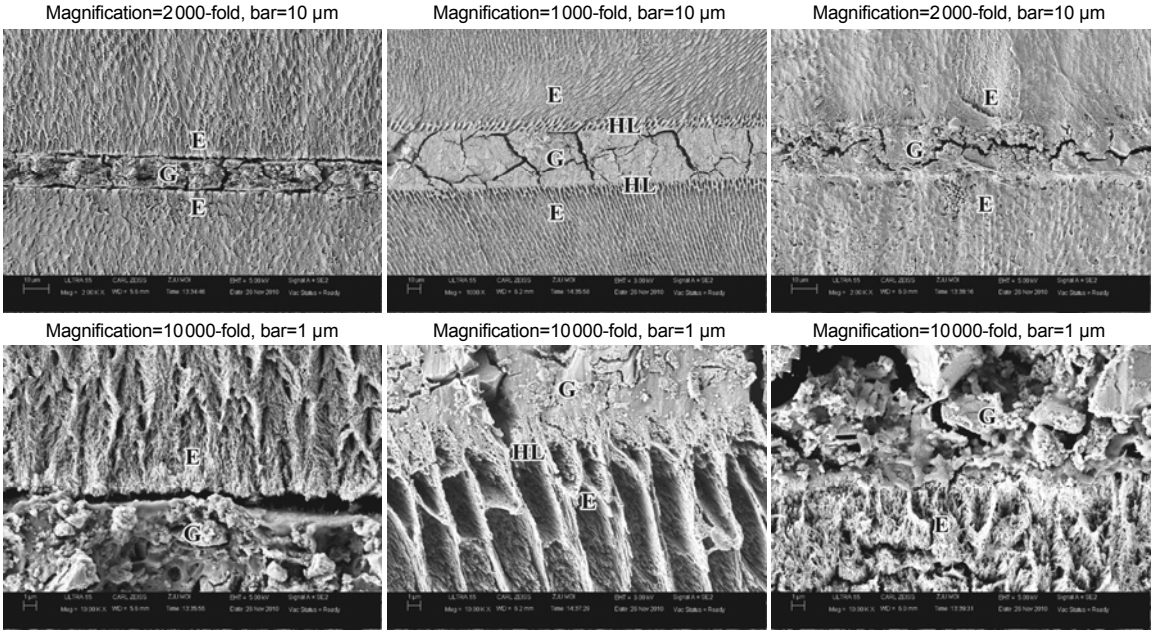


Fig. 4 Cross-sectional SEM view at the enamel-cement (Fuji I) interfaces

(a) The continuous micro-gaps were found between the polished enamel and the cement, and the hybrid layer was not visible; (b) Thick tag-like extensions penetrating into the acid-etched enamel substrate produced the thick hybrid layer between the enamel and the cement; (c) Conditioning with the liquid of the cement (Fuji I) resulted in a very thin hybrid layer between the enamel and the cement. G: glass ionomer cements; E: enamel; HL: hybrid layer

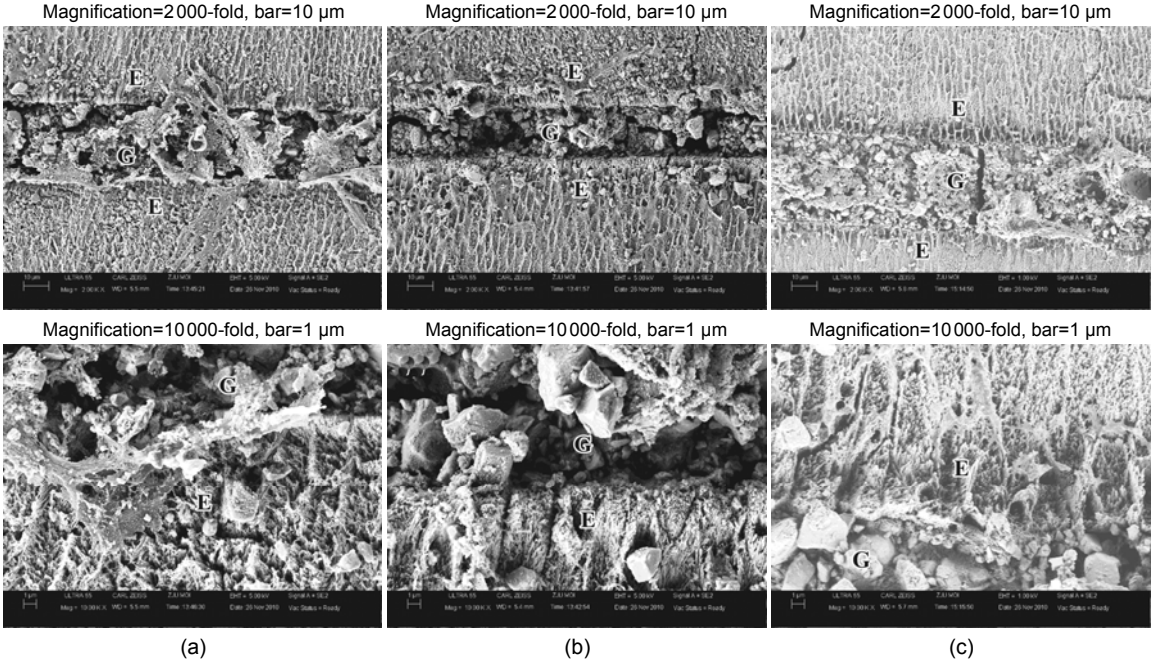


Fig. 5 Cross-sectional SEM view at the enamel-cement (Ketac Cem Easymix) interfaces

(a) The discontinuous micro-gaps were found between the polished enamel and the cement, and the hybrid layer was almost invisible; (b, c) The very thin hybrid layers were found between the enamel and the cement when the enamel surfaces were either acid-etched (b) or conditioned with the liquid of the cement (Ketac Cem Easymix) (c). G: glass ionomer cements; E: enamel

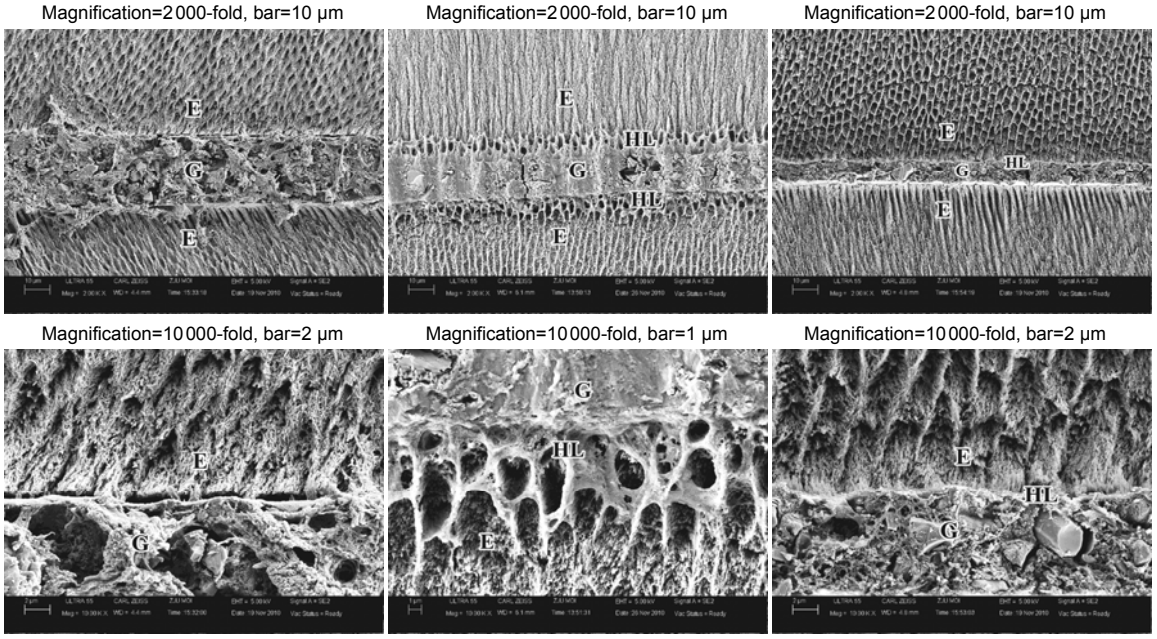


Fig. 6 Cross-sectional SEM view at the enamel-cement (Fuji Plus) interfaces

(a) The discontinuous micro-gaps with an invisible hybrid layer were found at the enamel-cement interface; (b) Typical resin tags penetrated into the acid-etched enamel substrate with network structures at the cement-enamel interfaces; (c) Polyalkenoic acid conditioning with the liquid of Fuji Plus produced the very thin hybrid layer. G: resin-modified glass ionomer cements; E: enamel; HL: hybrid layer

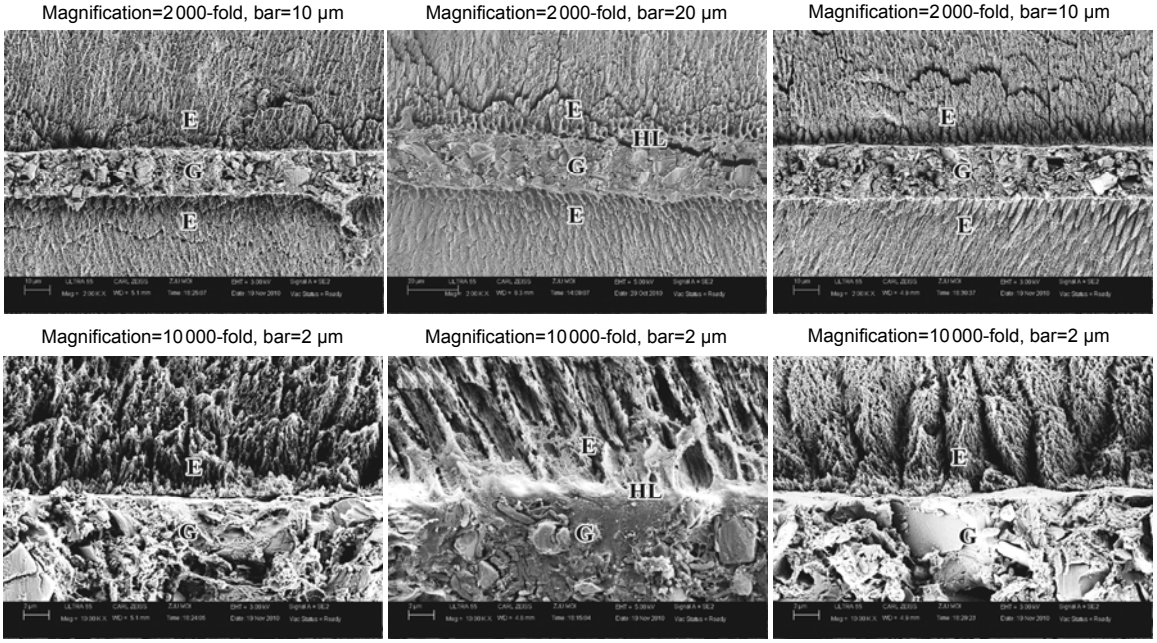


Fig. 7 Cross-sectional SEM view at the enamel-cement (RelyX Luting) interfaces

(a) A few micro-gaps with invisible hybrid layer were found at the enamel-cement interface; (b) Tag-like extensions penetrating into the acid-etched enamel substrate appeared as the visible hybrid layer at the enamel-cement interfaces; (c) The polyalkenoic acid conditioning with the liquid of RelyX Luting produced the very thin, continuous hybrid layer at the enamel-cement interfaces. G: resin-modified glass ionomer cements; E: enamel; HL: hybrid layer

Two matching pretreated enamel surfaces directly cemented together with GICs/RMGICs under finger pressure were chosen in this study, because dental prostheses are usually inserted onto or into prepared teeth under finger pressure and this method was also reported in a previous study (Altintas *et al*., 2008). The thickness of the cement layer in the present study was about 10–30 μm. This finding demonstrated that finger pressure is feasible and reliable for this study. In addition, fractography of most debonded specimens revealed cohesion failure of cements (Type 3 failure) due to low mechanical strengths of GICs/RMGICs. This helps to explain the reason that bond strength of GICs/RMGICs was low (Fajen *et al*., 1990; Wiltshire, 1994; Itoh *et al*., 1999).

4.1 Etching or conditioning

In the previous studies, the application of 37% phosphoric acid for 15 s to etching the enamel surfaces was recommended, because 15-s etching produced reliable, clinically acceptable enamel bond strengths (Gardner and Hobson, 2001; Glasspoole *et al*., 2002; Coutinho *et al*., 2006). Moreover, the extension of etching time would not increase the enamel bond strengths due to the excessive loss of the enamel substrate (Legler *et al*., 1990). Thus, 15-s etching was applied in this study.

GICs/RMGICs possess the twofold bonding mechanisms to tooth hard tissues via micromechanical interlocking and chemical bonding at the cement-enamel interfaces (Powis *et al*., 1982; Yoshida *et al*., 2000; Glasspoole *et al*., 2002; Fukuda *et al*., 2003; Coutinho *et al*., 2006). In the present study, 37% phosphoric acid etching significantly improved enamel bond strengths of GICs/RMGICs. This is in complete agreement with the previous reports (Cortes *et al*., 1993; Bishara *et al*., 2000a; Glasspoole *et al*., 2002; Cacciafesta *et al*., 2003; Coutinho *et al*., 2006). Cacciafesta *et al*. (2003) found a substantial increase in bond strength to enamel with Fuji Ortho LC, when the enamel was etched with 37% phosphoric acid in comparison to the enamel conditioned with 10% (w/v) polyacrylic acid. Coutinho *et al*. (2006) demonstrated that the pretreatments of the enamel surfaces with 37% phosphoric acid and 25% (w/v) polyalkenoic acid could significantly increase the enamel bond strengths for both Fuji BOND LC and Fuji LC, but acid-etching yielded much stronger enamel bond strengths than conditioning. Contrarily, Valente *et al*. (2002) showed that 37% phosphoric acid etching for 30 s produced enamel bond strengths for Fuji Ortho LC no stronger than either 25% phosphoric acid etching or 10% polyacrylic acid conditioning. These controversial findings might be related to different surface treatments, different modes of measurement, different cements used, and different operators (Cortes *et al*., 1993; Bishara *et al*., 2000a; Glasspoole *et al*., 2002; Valente *et al*., 2002; Cacciafesta *et al*., 2003; Coutinho *et al*., 2006; Jiang *et al*., 2010).

The role of phosphoric acid etching involves the effective removal of the smear layer (Fig. 3b) and provides good wetting of the surface with GICs, which can chemically interact with the underlying enamel (Torii *et al*., 2002; Brauchli *et al*., 2010). In addition, phosphoric acid etching produces micro-porosities in the enamel substrate and increases the surface area (Fig. 3b) for chemical bonding and micromechanical interlocking (Figs. 4b, 6b, and 7b) (Glasspoole *et al*., 2002; van Meerbeek *et al*., 2010). The SEM findings in this study for the first time revealed that certain cements could infiltrate the partially demineralized enamel substrate to form the typical micromechanical interlocking (Figs. 4b, 6b, and 7b), while other cements could not (Fig. 5b). The discrepancies might result from the permeability and acidity of the cements themselves. Furthermore, the typical micromechanical interlocking was not directly associated with the enamel bond strengths (Figs. 4b, 6b, and 7b; Table 2) in this study. In other words, the visible hybrid layer at the enamel-cement interface did not possess a stronger enamel bond strength than the invisible hybrid layer. This finding might be attributed to the sub-micro- or nano-scale hybrid layer at the enamel-cement interfaces, namely, the nano-interaction zone (NIZ) (Koshiro *et al*., 2006). Thus, micro- and nano-mechanical interlocking might play an important role in the presence of polymer tags penetrating into the micro-porosities of the enamel surfaces treated with etch-and-rinse adhesives or self-etch adhesives (Hannig *et al*., 2002). This was the reason that the acid-etching could substantially increase the enamel bond strengths of GICs/RMGICs in the present study.

In order to maximize the chemical bonding on the enamel surfaces, a diluted polyalkenoic acid conditioner was recommended to treat the enamel

surfaces prior to bonding (Coutinho *et al*., 2006). The liquids of GICs/RMGICs are polyalkenoic acid in the majority of common cements, while only a few liquids of common cements, such as Ketac Cem Easymix, contain tartaric acid (Table 1). However, conditioning with liquids of cements either did not increase the enamel μTBS, or even reduced the enamel μTBS in comparison with the control in the present study. Moreover, the SEM findings revealed that the hybrid layers were thin or even invisible when the polished enamel surfaces were conditioned with the liquids of the cements (Figs. 4c, 5c, 6c, and 7c).

Compared with 37% phosphoric acid, polyalkenoic acid in the conditioner might not be at a sufficiently high concentration to facilitate cleaning and wetting the enamel (Valente *et al*., 2002). The SEM findings in this study demonstrated that polyalkenoic acid conditioning, using liquids of Fuji Plus and RelyX Luting, could not effectively remove the polishing scratches on the smear layer (Figs. 3e and 3f). Oddly, conditioning with the liquids of Fuji I/Easymix could completely remove the smear layer with exposures of the prisms and the interprisms, but still did not increase the enamel bond strengths (Table 2; Figs. 4c and 4d). The tartaric acid used in the liquid of Ketac Cem Easymix (Table 1) could simultaneously decalcify and adhere to the enamel surface (Fu *et al*., 2005), but it could not increase the enamel bond strengths of the cement Ketac Cem Easymix. Still, these findings could be explained by the fact that various minerals or salts resulting from the chemical interaction of polyalkenoic acid or carboxylic acid with the enamel HAp, possibly deposited onto the enamel surfaces, and interfered with the intimate contact between the cements and the enamel substrate. In other words, the HAp at the enamel surfaces was saturated with the ionic bond provided by the carboxyl groups of polyalkenoic acid or carboxylic acid in the conditioner. This might block the further formation of the ionic bond between the enamel HAp and the carboxyl groups of GICs/RMGICs themselves, suggesting unfavorable surfaces for the effective bonding (Kanca, 1993; Valente *et al*., 2002).

Some researchers have advocated for there being no need to etch or condition tooth hard tissues before bonding procedures because GICs/RMGICs can self-adhere to tooth hard tissues (Wilson and Kent, 1972; Yoshida *et al*., 2000; 2001; Fukuda *et al*., 2003), and especially, RMGICs can bond to the enamel effectively (Silverman *et al*., 1995; Cacciafesta *et al*., 1998; 1999). However, others have insisted that enamel surface pretreatments could improve the bond strengths (Powis *et al*., 1982; Cortes *et al*., 1993; Attin *et al*., 1996; Bishara *et al*., 2000a; 2000b; Glasspoole *et al*., 2002; Valente *et al*., 2002; Cacciafesta *et al*., 2003; Coutinho *et al*., 2006). Interestingly, the findings in the present study revealed that the polished enamel surfaces produced much stronger, or at least, no weaker enamel μTBS than the conditioned enamel surfaces. This might be the reason why some researchers assert that it is needless to condition or etch the enamel surfaces before the bonding procedure. However, the acid-etched enamel surfaces resulted in significantly stronger enamel bond strengths than either the polished or the conditioned enamel surfaces in this study. According to our limited study, it is strongly recommended to use phosphoric acid to etch the enamel margins/surfaces before the cementation of dental fixed prostheses to improve the bond strengths. The integrity and bond durability of prostheses might be increased, with better bond performance (Erickson *et al*., 2009).

4.2 RMGIC vs. GIC

RMGICs were invented to overcome the disadvantages of GICs, such as low early mechanical strength and moisture sensitivity (Nicholson and Czarnecka, 2008; Yelamanchili and Darvell, 2008). As expected, the enamel bond strengths of RMGICs measured in this study were greater than those of GICs, regardless of etching or conditioning. RMGICs were able to bear more acidic challenges than GICs when they were briefly attacked by HCl solution (Figs. 6 and 7). The infiltration of the monomers of RMGICs into the enamel substrate and the monomers themselves, could play a role in the improvement of the bond strengths (Tanumiharja *et al*., 2000; Chitnis *et al*., 2006; van Landuyt *et al*., 2007).

Interestingly, the liquids of RMGICs in the present study possessed a weaker ability to condition enamel surfaces than those of GICs (Figs. 3e and 3f). This implies that the etching ability of polyalkenoic acid in RMGICs is much weaker than that of tartaric acid in Ketac Cem Easymix and polyacrylic acid/some polycarboxylic acid in Fuji I (Table 1). Whether the polished enamel surfaces were etched,

conditioned or not, RMGICs yielded much stronger enamel bond strengths than GICs. Thus, the enamel bond strengths of the cements do not rely on the etching patterns. They depend on the mechanical strengths of the cements themselves. The variations in the bond strengths of the cements were probably related to their individual material compositions rather than their bonding mechanisms (Triana *et al*., 1994; Glasspoole *et al*., 2002; Coutinho *et al*., 2006; Yelamanchili and Darvell, 2008). Phosphoric acid-etching can increase the enamel μTBS of GICs/RMGICs, but conditioning with the liquids of the cements does not increase the enamel μTBS of GICs/RMGICs.

5 Conclusions

Phosphoric acid etching significantly increases the enamel μTBS of GICs/RMGICs. Hence, it is recommended to etch enamel margins/surfaces before cementation of dental prostheses such as inlays or onlays using GICs/RMGICs. RMGICs provide stronger enamel bond strength than GICs, regardless of etching or not. Conditioning with the liquids of GICs/RMGICs does not increase enamel bond strength.

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Compliance with ethics guidelines

Ling ZHANG, Tian TANG, Zhen-liang ZHANG, Bing LIANG, Xiao-miao WANG, and Bai-ping FU declare that they have no conflict of interest.

All institutional and national guidelines for the care and use of laboratory animals were followed.

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