Shear Bond Strengths of Four Resin Bonding Systems to Two Silica-based Machinable Ceramic Materials

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The purpose of this study was to evaluate the bond strength between four bonding systems (GC Ceramic Primer and Linkmax HV (CP+LM), Clapearl Bonding Agent and Clapearl DC (CBA+CL), Clearfil Mega Bond Porcelain Bonding kit and Panavia F2.0 (MB+PF), and RelyX Ceramic Primer and RelyX ARC (RC+RA)) and two machinable ceramics (Vitablocs Mark II, VMII and GN-I ceramic block, GNI). Shear bond strength was determined after 24-hour immersion in water or after thermocycling of 20,000 cycles. It was found that the post-thermocycling bond strength of each bonding system to VMII was significantly higher than that to GNI. VMII showed no significant differences between CP+LM, MB+PF, and CBA+CL − the values of which were higher than that of RC+RA. Regarding GNI, CP+LM showed the greatest bond strength after thermocycling among the four bonding systems used. It was concluded that the crystalline phase of the ceramics used might have an effect on bond strength.

Key words : Machinable ceramic, Bonding system, Shear bond strength

INTRODUCTION

Patients usually desire natural-looking, tooth-colored restorations in prosthodontic treatment. With advancements in computerized manufacturing technology and adhesive bonding, the use of ceramic materials for esthetic dental restorations has increased substantially. Representative dental computer-aided design and computer-aided manufacturing (CAD/CAM) systems are the Duret system (Duret CAD/CAM, Henson International, Los Angeles, CA, USA)[1,2], Cerec system (Siemens AG, Bensheim, Germany)[3], GN-I system (GC Corp., Tokyo, Japan)[4], and DentiCAD system (BEGO, Bremen, Germany; DentiCAD USA, Waltham, MA, USA)[5,6]. A newer copy milling system (The Procera, Nobel Biocare Inc., Götebarg, Sweden) has also been developed and marketed.

The conventional sintered porcelain has the problem of sintering shrinkage during firing. On the other hand, CAD/CAM restorations are machined from homogenous and factory-standardized dental ceramic blocks. The microstructure of ceramics does not change with firing, fusion, or solidification, thereby retaining the original physical properties of the material. Silicon oxide glass ceramics, glass-alumina ceramics, glass-alumina-zirconia ceramics, alumina ceramics, and zirconia ceramics are used as machinable ceramic blocks.

The bonding of composites to ceramic materials plays an important role in dentistry[7−10]. Resin cements are used to lute conventional metal crowns, fixed partial dentures, ceramic crowns or veneers, and to repair fractured metal-ceramic restorations. The use of resin luting agents has the advantage of stress distribution, thereby preventing crack initiation[11]. It should be highlighted that a strong and durable bond between the tooth and ceramic restoration is indispensably important for extending both tooth life and restoration serviceability.

It has been reported that the bond strength between resin cements and silica-rich porcelains was improved with surface-modified porcelain and silane coupling agent[12−15]. Further, dual-cured resin cements were found to exhibit higher early bond strength to ceramic restorations rather than chemical-cured resin cements, and were able to maintain bond strength and durability[16]. Both Vitablocs Mark II (Vita Zahnfabrik, Bad Säckingen, Germany) and GN-I ceramic block (GC Corp., Tokyo, Japan) contain silicon oxide. Vitablocs Mark II is a conventional feldspathic ceramic with fine-grained particles[17], while GN-I ceramic block is a leucite glass ceramic. Previous research showed that a ceramic primer − consisting of separate solutions of an acidic monomer and a silane coupling agent − strongly coupled dual-cure resin cements to silicon oxide ceramics[18−20]. However, limited information is available on bonding to machinable silica-based ceramics. The purpose of this study, therefore, was to evaluate the durability of bond strength between four adhesive bonding systems and two machinable silica-based ceramics.

MATERIALS AND METHODS

Preparation of ceramic specimens
The materials and codes used in the present study are listed in Table 1. Two machinable silica-based glass ceramic blocks (VMII and GNI) were selected as the adherend substrate materials. Two ceramic specimens, a rectangular disk (10 × 8 × 2.5 mm) and a round disk (6 mm in diameter × 2.5 mm thick), were fabricated from the two ceramic blocks with a low-speed cutting saw (Isomet, Buehler Ltd., Lake Bluff, IL, USA). The ceramic surface was sanded with ♯600, ♯800, ♯1,000, and ♯1,200 silicon carbide papers and cleaned ultrasonically in 99.5% acetone for five minutes. The surfaces were treated with 40% phosphoric acid gel (Kuraray Medical Co. Ltd., Osaka, Japan) for five seconds, and then rinsed with water. A piece of polyethylene tape with a circular hole of 4-mm diameter was positioned on the surface of the rectangular disk specimen to define the bonding area.

Bonding procedure
Ceramic specimens were bonded together with one of the four resin bonding systems. Each ceramic primer was used according to the manufacturer's directions. With CP+LM, specimens were treated with CP, followed by air-drying and then bonded with LM. With CBA+CL, specimens were treated with CBA, followed by air-drying and then bonded with CL. With MB+PF, specimens were treated with MB, followed by air-drying and then bonded with PF. With RC+RA, specimens were treated with RC, followed by air-drying and then bonded with RA. A purpose-designed sample holder was used to control the film thickness of the luting agent at $50\,\mu$m. Excess luting agent was removed, and then the bonded specimens were irradiated with a visible light curing source (Quick Light, J. Morita Corp., Kyoto, Japan) for 40 seconds.

Shear bond strength test
Ten specimens were fabricated for each combination of two ceramics and four bonding systems. A total of 80 bonded specimens were randomly assigned to eight test groups. Each group was divided into two subgroups of five specimens each, with and without thermocycling. For thermocycling test, 40 specimens

Table 1 Materials assessed

Material	Code	Composition	Manufacturer	Batch No.	
Primer / bonding agent					
GC Ceramic Primer (2 liquid)	CP	Ethanol, silane coupler, methylmethacrylate, UDMA, organic acid	GC Corp., Tokyo, Japan	A liquid	0212181
				B liquid	0212181
Clapearl Bonding Agent (3 liquid)	CBA	Ethanol, methacrylate monomer, Bis-GMA, HEMA, MDP, photoinitiator, chemical initiator, silane coupler	Kuraray Medical Co. Ltd., Osaka, Japan	Universal	353AM
				Catalyst	455AM
				Porcelain Bond Activator	00132A
Clearfil Mega Bond Porcelain Bonding kit (2 liquid)	MB	MDP, HEMA, water, photoinitiator, silane coupler, methacrylate monomer	Kuraray Medical Co. Ltd., Osaka, Japan	Primer	00452B
				Porcelain Bond Activator	00155A
Rely X Ceramic Primer (1 liquid)	RC	Silane coupler, ethanol, water	3M ESPE, Seefeld, Germany		3UK
Luting agent					
Linkmax HV	LM	UDMA, filler (fluoro-alumino-silicate glass), photoinitiator, chemical initiator	GC Corp., Tokyo, Japan	A paste	0207261
				B paste	0207261
Clapearl DC	CL	Bis-GMA, TEGDMA, filler (barium glass, silicate glass), photoinitiator, chemical initiator	Kuraray Medical Co. Ltd., Osaka, Japan	A paste	0033BD
				B paste (Universal)	0027CC
Panavia F2.0	PF	MDP, methacrylate monomer, filler (barium glass, silicate glass), NaF, photoinitiator, chemical initiator	Kuraray Medical Co. Ltd., Osaka, Japan	A paste	00066A
				B paste (Brown)	00033A
Rely X ARC	RA	Bis-GMA, TEGDMA, dimethacrylate polymer, zirconia/silica glass (67.5wt%), photoinitiator, chemical initiator	3M ESPE, Seefeld, Germany		CYEA
Ceramic					
Vitablocs Mark II	VMII	Al_2O_3, SiO_2, Na_2O, K_2O feldspar	Vita Zahnfabrik, Bad Säckingen, Germany		
GN-I ceramic block	GNI	Al_2O_3, SiO_2, Na_2O, K_2O leucite	GC Corp., Tokyo, Japan		

UDMA: urethane dimethacrylate; Bis-GMA: bisphenol-A-diglycidylether dimethacrylate; MDP: 10-methacryloxydecyl dihydrogen phosphate; HEMA: 2-hydroxyethyl methacrylate; TEGDMA: triethylene glycol dimethacrylate

were stored in water at 37℃ for 24 hours, followed by 20,000 thermocycles between water baths held at 4℃ and 60℃ by means of a thermocycling apparatus (Rika-Kogyo, Hachioji, Japan) with a dwelling time of one minute in each bath (TC 20,000). As for the other 40 specimens, they were subjected to water storage at 37℃ for 24 hours without thermal cycling (TC 0).

All the bonded specimens were embedded in an acrylic resin mold and arranged in an ISO/TR 11405 shear testing jig (Wago Industrial Ltd., Nagasaki, Japan). Shear load was applied to bonded round disk specimens with a universal testing machine (AGS-10kNG, Shimadzu Corp., Kyoto, Japan) at a crosshead speed of 0.5 mm/min. Bond strength data were then analyzed by three-way analysis of variance (ANOVA), in which the resin bonding system, ceramic type, and number of thermal cycles were independent factors. Duncan's new multiple range test with $P < 0.05$ was also used to compare the mean values of five specimens.

Failure mode analysis
Debonded ceramic surfaces were observed through an optical microscope (SMZ-10, Nikon Corp., Tokyo, Japan) at $30\times$ magnification to assess bond failure. Failure modes were categorized as: adhesive failure at ceramic-luting agent interface (A); adhesive failure at ceramic-luting agent interface with crack propagation inside the ceramic (B); ceramic cohesive failure (C); complex A and C, or complex B and C (D).

RESULTS

Table 2 shows the means and standard deviations of shear bond strength together with the bonding failure modes. Three-way ANOVA results revealed that bond strength was significantly influenced by the following factors: bonding system ($p < 0.0001$), ceramic type ($p < 0.0001$), and thermocycling ($p < 0.0001$). The interactions were also significant in all combinations of the three factors.

No significant differences were observed between VMII and GNI ceramic blocks at 0 thermocycles. After 20,000 thermocycles, the bond strengths to GNI were significantly lower than those to VMII. As for VMII/CBA+CL, VMII/MB+PF, and VMII/CP+LM, they showed no significant differences between 0 and 20,000 thermocycles. When VMII was bonded with RC+RA, the bond strength was significantly reduced after 20,000 thermocycles. As for GNI, the bond strengths after 20,000 thermocycles were significantly lower than that after 0 thermocycles in each bonding system. In particular, GNI/CP+LM exhibited significantly higher bond strength than the other GNI groups after 20,000 thermocycles.

Evaluation of the failure modes after shear testing indicated that high bond strength groups showed mode C or D, while low bond strength groups tended to exhibit mode A. For VMII, mode C was found in most of the specimens. In contrast, GNI failed mostly with mode A.

DISCUSSION

The present study evaluated the shear bond strength and durability of four resin bonding systems to two machinable silica-based ceramics. Ceramics can be etched with hydrofluoric acid to provide micromechanical retention. However, to exclude the effect of micromechanical retention, neither mechanical modification of ceramic surface nor hydrofluoric acid etching was employed in this study.

VMII/CBA+CL exhibited the greatest bond strength both before and after thermal cycling. Moreover, most VMII specimens showed cohesive failure within the ceramic material, indicating that a strong bonding was established with VMII. After

Table 2 Shear bond strengths and failure modes of four resin bonding systems bonded to VMII or GNI

Group Name	0 thermocycles					20,000 thermocycles				
	Mean (SD)* [MPa]	Number of specimens corresponding to each failure mode**				Mean (SD)* [MPa]	Number of specimens corresponding to each failure mode**			
		A	B	C	D		A	B	C	D
VMII/RC+RA	43.6 (4.8)[d,e]	1	3	1	0	29.6 (5.6)[b]	5	0	0	0
VMII/CBA+CL	45.3 (6.7)[e]	0	0	5	0	46.9 (7.3)[e]	0	0	5	0
VMII/MB+PF	42.6 (8.4)[d,e]	0	1	1	3	46.1 (3.6)[e]	0	3	2	0
VMII/CP+LM	45.9 (1.6)[e]	0	3	2	0	42.2 (2.3)[d,e]	0	1	4	0
GNI/RC+RA	36.6 (6.3)[c,d]	2	2	1	0	1.6 (1.5)[a]	5	0	0	0
GNI/CBA+CL	41.4 (8.1)[d,e]	0	0	4	1	3.0 (1.1)[a]	5	0	0	0
GNI/MB+PF	42.0 (6.2)[d,e]	2	2	0	1	1.6 (2.7)[a]	5	0	0	0
GNI/CP+LM	41.9 (3.8)[d,e]	1	2	2	0	30.5 (3.7)[b,c]	1	3	1	0

*Same letters at both thermocycles indicate that the values are not statistically different ($p > 0.05$).
**A: Adhesive failure at ceramic-luting agent interface; B: Adhesive failure at ceramic-luting agent interface with crack propagation inside ceramic;
  C: Ceramic cohesive failure; D: Complex A and C, or complex B and C.

20,000 thermocycles, GNI showed significantly lower values than VMII. The mode of failure was mostly adhesive failure between GNI ceramic and resin cement. These findings suggested that GNI was inferior to VMII in adhesive bonding. In the clinical situation, additional micromechanical retention may be needed when luting dental restorations fabricated with GNI.

VMII is a conventional feldspathic ceramic with fine-grained particles[17]. GNI is a leucite glass ceramic. The chemical formula of feldspar is $K_2O \cdot Al_2O_3 \cdot 6SiO_2$, while that of leucite crystal is $K_2O \cdot Al_2O_3 \cdot 4SiO_2$. It has been reported that a silane coupling agent improved the bond strength between a machinable ceramic and some resin cements[18,20,21]. This was because the silane coupling agent reacted with Si-OH groups on the ceramic surface[22]. Therefore, it was speculated that the difference in crystalline phase between GNI and VMII — arising from the difference in their SiO_2 contents — influenced the bond strength.

With each ceramic material, GNI or VMII, bond durability varied among resin bonding systems. LM contained urethane dimethacrylate (UDMA). On the other hand, CL and RA contained bisphenol-A-diglycidylether dimethacrylate (Bis-GMA) and triethylene glycol dimethacrylate (TEGDMA). Water sorption values of poly-TEGDMA, poly-Bis-GMA, and poly-UDMA were 6.33, 2.93, and 2.59% respectively[23]. However, some resin cements showed a wide variation in the magnitude of water sorption at equilibrium[24]. Nonetheless, regardless of the degree of water sorption, water most probably diffused to the interface between resin cement and ceramic surface through the resin matrix. Further, some bonding agents contained 2-hydroxyethyl methacrylate (HEMA), Bis-GMA, or UDMA which exhibit high water sorption[25]. The degree of water sorption of some commercial bonding agents containing HEMA varied from 5% to 11.6%[26]. Therefore, chemical compositions of resin cements, primers, and bonding agents may influence the hydrolysis of the siloxane bond at the interface.

The primers and bonding agents used are classified as either unhydrolyzed multi-bottle primers (CP, CBA, and MB) or pre-hydrolyzed single-bottle primer (RC). Both pre-hydrolyzed single-bottle primers and unhydrolyzed multi-bottle primers use water or different organic acids to activate the silane coupling agent[27,28]. Although activation of silane coupling agent contributes to ceramic bonding, this may limit the shelf life of the pre-activated silane coupling agent in the single-bottle primer.

In conclusion, GNI showed significantly lower bond strength than VMII after 20,000 thermocycles. This might be due to the difference in crystalline phase: feldspar group of VMII *versus* the leucite group of GNI. With GNI, use of CP+LM bonding system seemed to yield significantly higher bond strength than the other bonding systems evaluated (CBA+CL, MB+PF, and RC+RA).

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