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Effect of surface condition of dental zirconia ceramic (Denzir) on bonding

(Running title: SURFACE CONDITION OF ZIRCONIA BONDING)

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Abstract

Yttria partially-stabilized zirconia (YPSZ) ceramics are suitable for dental and medical use because of their high fracture toughness and chemical durability. The purpose of this study was to examine the bonding of a dental YPSZ ceramic (Denzir). After being subjected to various surface treatments the Denzir specimens were bonded to each other using an adhesive resin composite, glass ionomer or zinc phosphate cement. The bonding strength was then determined by the shearing test. No significant difference ($p > 0.05$) was observed between SiC- and Al_2O_3-blasted specimens. In all surface treatments, the shear bonding strength significantly ($p < 0.05$) increased in the order of adhesive resin composite cement > glass ionomer cement > zinc phosphate cement. Silanization with methacryloxy propyl trimethoxysilane slightly increased the bonding strength of the adhesive resin composite.

Keywords: Zirconia, Bonding strength, Surface treatment

Introduction

In recent years the demand for dental restorations with more aesthetic and biocompatible properties has increased and various ceramics have been widely studied and applied as materials for dental restorations[1-6]. Yttria partially stabilized zirconia (YPSZ) is a ceramic that exhibits superior fracture toughness and chemical durability compared to other ceramics[1, 2]. Because of these suitable properties, YPSZ has been extensively employed for medical and dental uses[2-5]. However, manufacturing of dental restorations requires complicated machining of various shapes and dimensions and it is difficult to achieve accurate machining. Through recent progress in the CAD/CAM-technique, YPSZ ceramics can, however, now be employed for complicated dental restorations such as inlays, copings and fixed partial denture (FPD) frameworks[6-10]. Dental restorations using YPSZ are made either by milling enlarged restorations out of homogenous ceramic green-body blanks of zirconia, which are then sintered and shrunk to the desired final dimensions[7], or by milling the restorations directly with the final dimensions out of highly dense sintered prefabricated YPSZ blanks, called hot isostatic pressed (HIPed) zirconia blanks[6, 8-10].

In an earlier report, the authors determined the cytotoxicity and bonding property of HIPed YPSZ[11]. In that study[11], HIPed YPSZ showed no cytotoxicity and exhibited high bonding to glass ionomer cement, but relatively low bonding strength to adhesive resin composite cement [11]. Kern et al., however, reported that a durable bond to YPSZ was achieved by using a phosphate monomer containing resin composite bonding cement[12]. In their study[12], as in most of the earlier studies evaluating the bond strengths of various cementing agents to YPSZ ceramics[e.g. 13-19], specimens made out of homogenous ceramic green zirconia were used. Since HIPed YPSZ blanks have been successfully employed in the fabrication of dental all-ceramic crowns and FPDs[8-10], the bonding properties of this type of zirconia ceramic are of interest. However, in a survey of the literature, only 1

article[11] was found that addressed the bonding properties of HIPed YPSZ. The purpose of this study, therefore, was to evaluate the bonding properties of various bonding cements to HIPed YPSZ and the effects of surface roughness and pretreatments on bonding strength.

Materials and methods

Twenty HIPed YPSZ (Denzir, Cad.esthetics AB, Skellefteå, Sweden) specimens were machined into a rectangular shape (14 mm x 14 mm x 5 mm) using the Cad.esthetics CAD/CAM system (Cad.esthetics AB). Some of these specimens were then sandblasted with 70 μm Al_2O_3 (Hi-Alumina, Shofu, Kyoto, Japan) and the remainder with 125 μm SiC powder (Carborundum, Shofu, Kyoto, Japan) with 0.3 MPa for 30 sec at a distance of 10 mm. Thereafter, SiC-blasted specimens were etched with a phosphoric acid gel (K-etchant gel, Kuralay, Kurashiki, Japan) for 10 seconds. In addition, the etched specimens intended to be cemented with an adhesive resin composite cement were then silanized by either: *i*) treatment with Clearfil Porcelain Bond Activator (Kuraray, Kurashiki, Japan) mixed with Clearfil Linerbond IIS (Kuraray, Kurashiki, Japan) for 10 sec in accordance with the manufacturers' instructions, or *ii*) heating with 10% methacryloxy propyl trimethoxysilane (MPTS, Tokyo Kasei Kogyo, Tokyo, Japan)-toluene (dehydrated) solution at 60°C for 1 hour and drying in a vacuum.

The cements used for the bonding test were *i*) a zinc phosphate cement (Elite Cement, GC, Tokyo, Japan), *ii*) a glass ionomer cement (Fuji I, GC, Tokyo, Japan) and, *iii*) an adhesive resin composite cement (Panavia 21, Kuraray, Kurashiki, Japan). Using one of the above-mentioned cements, the surface-treated specimens were bonded to corresponding specimens with a similar surface condition. The bonding area was 14mm x 5mm. The bonded specimens were then stored at 37°C, 100% RH for 1 hour. Seven replicates were prepared for each condition. The abbreviations of surface treatments and cements are tabulated in Table 1.

Table 1

The surface profile and mean surface roughness (Ra) were determined using a

surface profilometer (Surfcom 2000, Tokyo Seimitsu Co.Ltd., Tokyo, Japan).

The bonding strength was determined by the shearing test method using a universal testing machine (Model 4204, Instron, Canton, U.S.A.). The apparatus for shear test is shown in Fig. 1. The shear force was applied at cross-head speed of 0.5mm/sec. The results of the bonding test were analyzed statistically using ANOVA and Scheffe's test at a significance level of $p < 0.05$. After the bonding test, the YPSZ blocks were polished with emery paper (#80) until the attached cement layer was removed. Then, the polished blocks were surface treated again and used for the next bonding test.

Fig. 1

After the shear test, the fracture surfaces were analyzed using a scanning electron microscope (SEM) (S-2380, Hitachi, Tokyo, Japan). With an energy dispersed X-ray detector (EDX) (Genesis, EDAX Japan, Tokyo, Japan) the elemental distribution image of the observed surface was determined.

Results

Figure 2 shows the surface profiles and the mean surface roughness level (Ra) of Al_2O_3- and SiC-blasted and phosphoric acid-etched YPSZ specimens. The SiC-blasted surfaces had a clearly rougher profile than the Al_2O_3-blasted surfaces. The mean surface roughness (Ra) of the SiC-blasted surface was 0.86 μm, twice that of the Al_2O_3-blasted surface (0.43 μm).

Fig. 2

The shear bonding strengths of the Al_2O_3- and SiC-blasted and phosphoric acid-etched YPSZ specimens cemented with the three types of cement are shown in Fig. 3. In all surface treatments, the shear bonding strength increased in the order of adhesive resin composite cement > glass ionomer cement > zinc phosphate cement. Among the three types of cement, the bonding strength differed significantly ($p < 0.05$). Between the Al_2O_3- and SiC-blasted YPSZ specimens there was no significant difference ($p > 0.05$) in the shear bond strengths obtained. Thus, the bonding strength depended on the type of cement, and the surface roughness did not significantly affect the bonding strength.

Fig. 3

Figure 4 shows the effects on the bonding strength of surface treatments using phosphoric acid, PBA or MPTS in combination with the various cements. The significance of difference is tabulated in Table 2. In all surface treatments, the shear bonding strength increased in the order of adhesive resin composite cement > glass ionomer cement > zinc phosphate cement. Among the adhesive resin cement, The MPTS treatment showed highest bonding strength but their difference was in low significance ($0.1>p>0.05$).

Fig. 4

Table 2

Figure 5 shows the SEM and elemental distribution images of the fracture surfaces bonded with zinc phosphate, glass ionomer and adhesive resin composite

Fig. 5

cements. The surfaces of the zinc phosphate cemented specimens were clearly separated into a cement part (Zn rich area) and a YPSZ surface (Zr-rich area). The cement/YPSZ boundary was clear and interface fracture between the YPSZ and zinc phosphate cement was suggested. In contrast, the glass ionomer- and adhesive resin composite cement-bonded specimens showed complex, intricate surfaces, which implied partly cohesive fracture.

Discussion

In this study, the effects on the shear bond strength of various surface conditions of YPSZ in combination with various cements were studied. The bonding strength of YPSZ with two different surface roughnesses and three types of cement was tested. The different surface roughnesses studied did not influence the bonding strength, whereas the different types of silanization and cement used significantly affected the bonding strength of the adhesive resin composite cement. In all surface pretreatments, the shear bonding strength increased in the order of adhesive resin composite cement > glass ionomer cement > zinc phosphate cement.

In a previous report, the authors determined the bonding strength of YPSZ using threaded and tapered holes in a brass plate and YPSZ milled specimens.[11] In that study[11], the glass ionomer cement had the highest bonding strength, whereas no significant difference was seen between adhesive resin composite and zinc phosphate cements.[11] In contrast, in the present study the adhesive resin composite cement showed the highest bonding strength. Differences in the test methods are considered to be one possible cause for the different results obtained in the studies. In the previous study[11], the specimens and the brass plate were tapered. Therefore, the applied force probably included not only shearing force but also tensile force. In addition, when luted with the various cements used, the YPSZ specimens were placed manually in drilled tapered holes.[11] This may have resulted in cement layers whose thickness was not constant. With the present method, the flat surface of the YPSZ specimen was bonded to another surface with a similar surface condition, which should reproduce the bonding condition more consistently that the previous method. In the present study, the shearing force was applied in the direction perpendicular to the bonding plane. The present method should, therefore, more

accurately determine the bonding strength to YPSZ of the three types of cement studied.

The MPTS treatment with the adhesive resin cement showed the higher shear bonding strength than the other treatment, but their difference was in low significance ($0.1>p>0.05$). The reason for the difference of effectiveness between MPTS and PBA is considered to be as follows. In the PBA treatment, a mixture of PBA and the primer (Clearfil Linerbond IIS) was applied on the YPSZ surface and air-dried. In contrast, the MPTS treatment was carried out at 60^{o}C and drying was done in a vacuum after removal of the residual MPTS/toluene solution. Therefore, the formed surface layer would be thinner and more tightly bonded to the YPSZ surface than when using the PBA pretreatment. Thus, the results in the present study indicated that the adhesive resin composite cement in combination with MPTS pretreatment bonded more effectively to the YPSZ than in combination with the PBA treatment.

Values reported for the bonding strength of various adhesive resin-cementing agents to zirconia ceramic specimens in previous studies were between 1.7 MPa and 70.4 MPa.[e.g. 17, 20)] In most of the studies that addressed the bonding strength to zirconia ceramics, the specimens were made of homogenous ceramic green-body blanks of zirconia, and various bonding systems and pretreatments were used.[e.g. 13-19] In the current study, specimens of highly dense sintered prefabricated zirconia blanks, called hot isostatic pressed (HIPed) zirconia blanks, were used. In an earlier report on the bonding strength of HIPed YPSZ zirconia[11)], the mean values were 12 MPa for adhesive resin composite cement, 21 MPa for glass ionomer cement and 9 MPa for zinc phosphate cement. That is, the mean value of the resin composite cement was close to the value obtained in the current study, whereas the values obtained for the zinc phosphate and

glass ionomer cements were lower.

The effects of surface treatments on bonding to dental ceramics other than zirconia have also been presented in earlier studies.[12, 21-24)] The shear bonding strengths when using adhesive resin composite cements and silanated surfaces of a lithium disilicate glass-ceramic and feldspar ceramics have been reported to be 47 MPa for Empress2[12)] (Ivoclar Vivadent, Schaan, Lichtenstein), 18 MPa for Vita VMK 68[22)], 42 MPa for Cerec 2 Vitablock Mark II[23)] and 55 MPa for Vita Celay[24)] (all Vita products from Vita Zahnfabrik, Bad Säckingen, Germany). Aida et al. reported that the bonding strength of another feldspar ceramic (Laminabond Porcelain, Shofu, Kyoto, Japan) increased with silanization using commercial priming silane agents other than MPTS [21]. Those results partly contradict the results obtained in the present study. The reason could be differences in the silane treatment and the ceramics studied. In this context it should be noted that hydrofluoric acid etching of conventional silica-based ceramics usually improves the effect of silanization and adhesive bonding[12)], whereas dental zirconia ceramics are unetchable with hydrofluoric acid. However, it should also to be pointed out that it has been said that the phosphate ester group in the Panavia 21 resin composite directly bonds to metal oxides[26)] and bonds chemically to zirconia ceramics.[12)]

The reported bonding strengths of cements to dental ceramics, thus, vary within a wide range and assessment of their clinical significance is difficult. It has been suggested that 10-13 MPa is the minimum strength needed for clinical bonding[27, 28)] On the other hand, the in vitro bond strengths to acid-etched human dentin of various commercial resin composite bonding cements, which have been in clinical use for a relatively long time, are reported to range from 1.1 MPa to 14.8 MPa.[29)] For conventional zinc phosphate cement, Øilo[30)] reported a tensile bond strength to dentin of 0.6 MPa and Richardsson et al. 0.9 MPa[31)] Although those values seem to be very low, and are

considerably inferior to those suggested as the acceptable minimum strength for clinical bonding[27, 28], zinc phosphate cements have been successfully used clinically for a very long time to lute cast dental restorations. To assess the clinical performance of bonding systems, in vitro studies should, therefore, be supplemented with clinical studies with long follow-up times.

In most of the earlier studies dealing with zirconia specimens, various surface pre-treatments such as sandblasting or tribochemical methods for silicatizing the surfaces were used[e.g. 13-19] However, mechanical treatments of zirconia should be done with caution because it has been demonstrated that heat treatment, sandblasting and grinding can influence its mechanical properties[10, 32, 33] In a recently presented study by Sundh and Sjögren it was stated that the effect on the fracture resistance of zirconia depended on, among other things, the time the specimens were subjected to sandblasting.[10] This is probably because sandblasting treatment and/or grinding can induce compressive stresses and/or phase transformation on the surface, which increases the strength, but also can induce flaws and other defects that reduce strength. Thus, to find the best possible technique more studies are needed to determine the effects of surface treatments on the bond strength and mechanical properties of zirconia ceramics.

Conclusion

Within the limitations of this in vitro study the following conclusions were drawn:

1 The different surface roughnesses obtained between SiC- and Al_2O_3-blasted specimens did not significantly affect the bonding strength.

2 In all surface pretreatments, the shear bonding strength significantly increased in the order of adhesive resin composite cement > glass ionomer cement > zinc phosphate cement.

3 The adhesive resin composite cement bonds slightly better to YPSZ in combination with 10% methacryloxy propyl trimethoxysilane-toluene (dehydrated) solution than the other surface treatment.

Acknowledgements

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Figure Captions

Figure 1 Apparatus for the shear bonding test.

Figure 2 Surface profiles and mean surface roughness (Ra) of YPSZ specimens blasted with Al_2O_3 or SiC.

Figure 3 The shear bonding strength of Al_2O_3- and SiC-blasted and phosphoric acid-etched YPSZ in combination with the three cements studied.

Figure 4 The effect on the bonding strength to SiC-blasted and phosphoric acid-etched YPSZ specimens of various surface treatments in combination with the three cements studied.

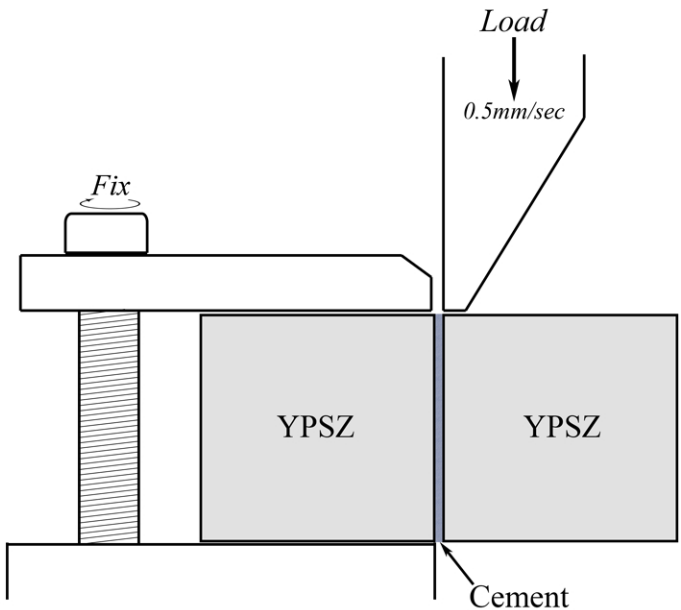
Figure 5 SEM and elemental distribution images of fractured surfaces bonded with various cements. (C: cement, Z: YPSZ)

Table 1 The abbreviations for surface conditions and cements.

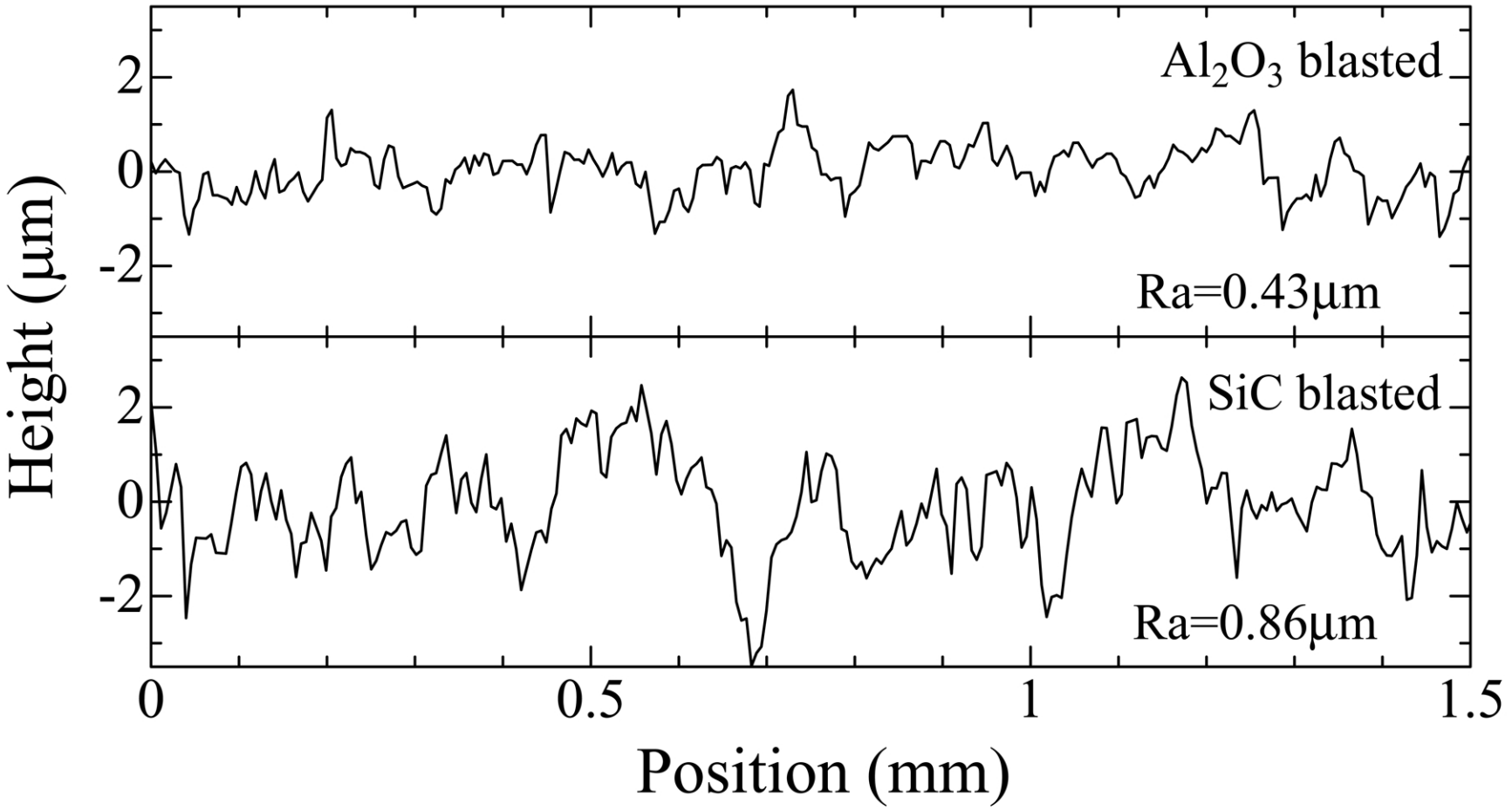
Abbrebiations	Cements or surface treatments
ZP	zinc phosphate cement
GI	glass ionomer cement
AR	adhesive resin composite cement
AB	Al_2O_3-blasted
SB	SiC-blasted
PE	phosphate gel-etched
PBA	treated with Clearfil Porcelain Bond Activator
MPTS	treated with 10% methacryloxy propyl trimethoxysilane-toluene solution

Table 2 Summary of statistical analysis of the bond strength of the SiC blasted specimens. Results of ANOVA supplemented with Scheffe's test. (n.s.: no significant difference, *: $p<0.05$, **: $p<0.001$)

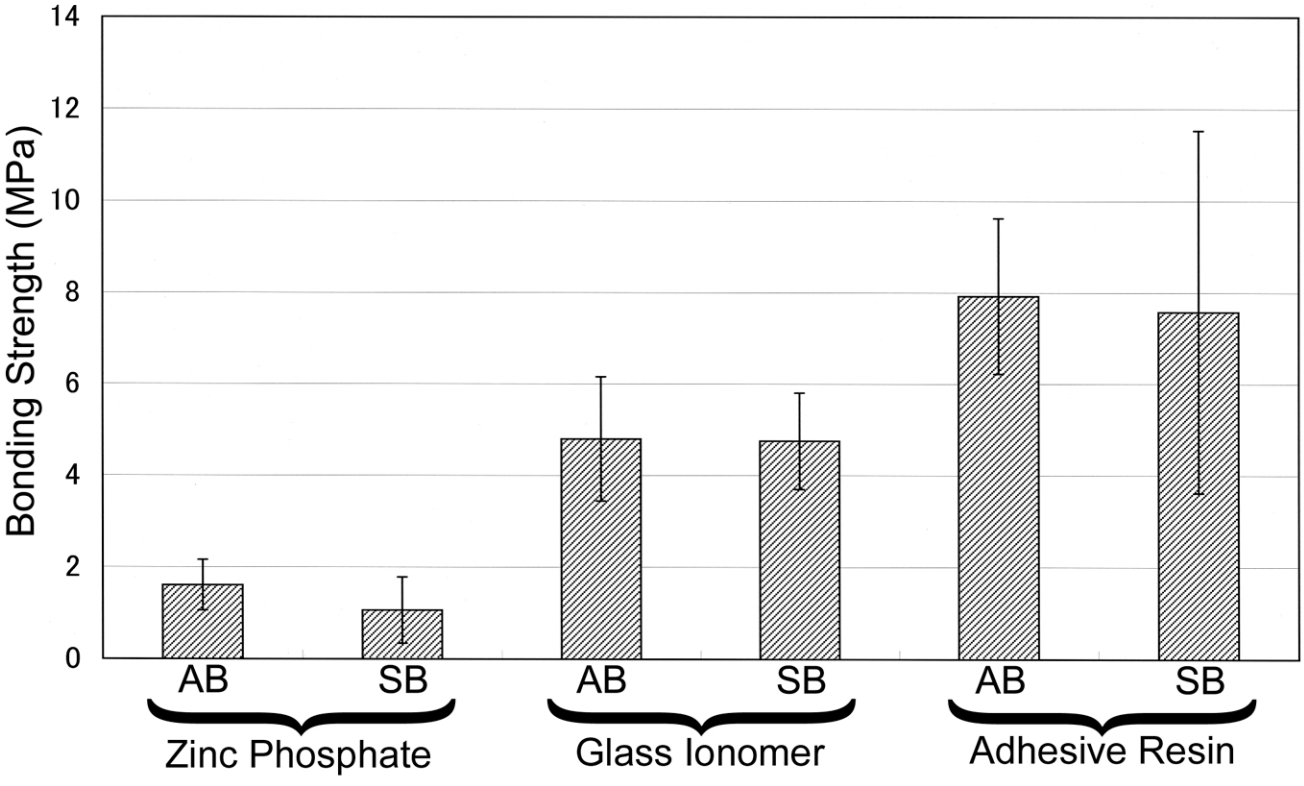
	Cements	ZP		GI		AR			
Cements	surface treatment	SB	SB PE	SB	SB PE	SB	SB PE	SB PE PBA	SB PE MPTS
ZP	SB								
	SB-PE	n.s.							
GI	SB	**	**						
	SB-PE	**	**	n.s.					
AR	SB	*	*	n.s.	n.s.				
	SB-PE	*	*	*	n.s.	n.s.			
	SB-PE-PBA	**	**	**	**	n.s.	n.s.		
	SB-PE-MPTS	**	**	**	*	n.s.	n.s.	n.s.	



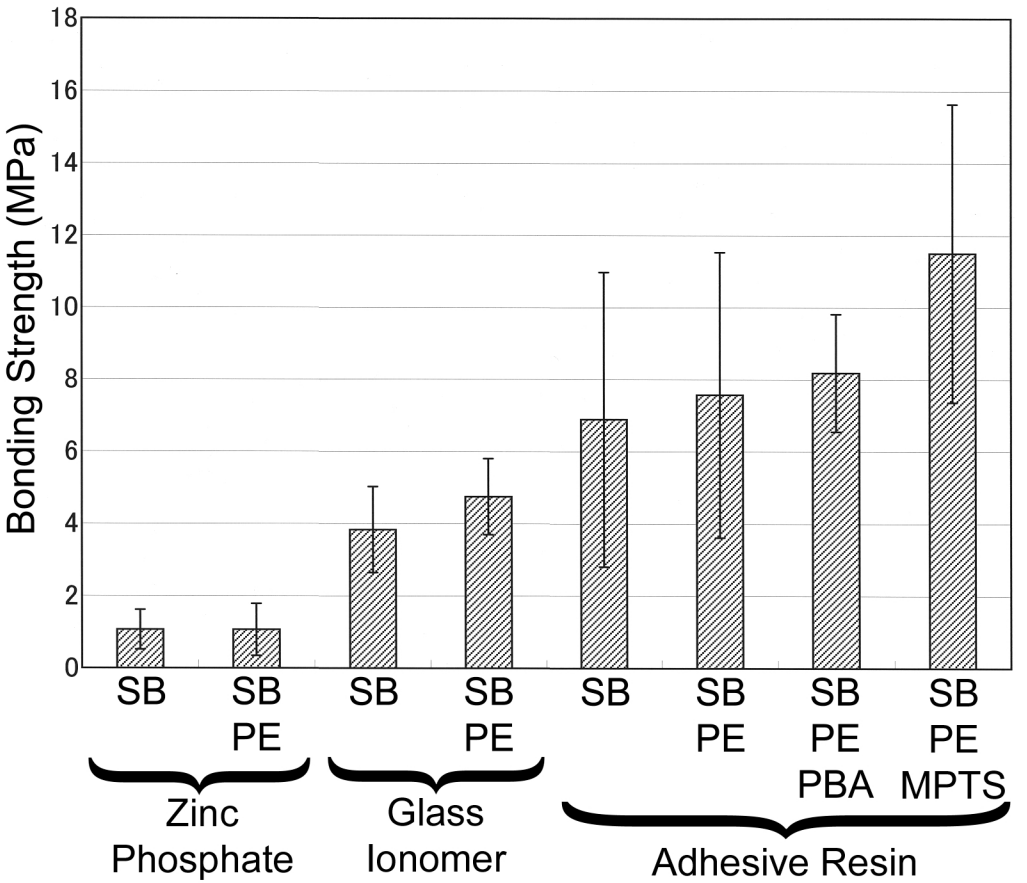
(Fig.1)


Al2O3 blasted
Ra=0.43μm
SiC blasted
Ra=0.86μm
Height (μm)
2
0
-2
0
0.5
1
1.5
Position (mm)

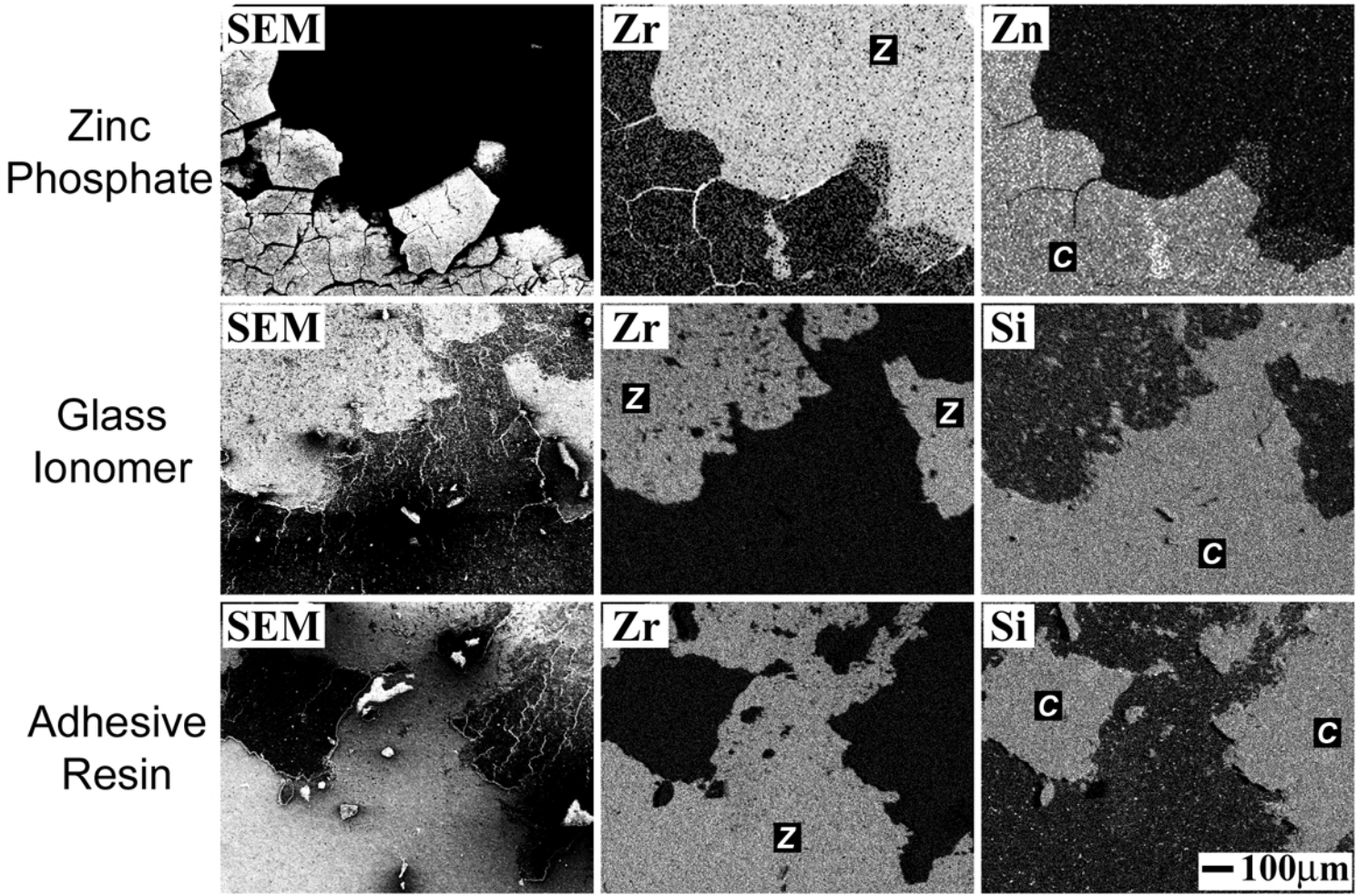
(Fig.2)


Bonding Strength (MPa)
14
12
10
8
6
4
2
0
AB
SB
AB
SB
AB
SB
Zinc Phosphate
Glass Ionomer
Adhesive Resin

(Fig.3)


Bonding Strength (MPa)
18
16
14
12
10
8
6
4
2
0
SB
SB PE
SB
SB PE
SB
SB PE
SB PE PBA
SB PE MPTS
Zinc Phosphate
Glass Ionomer
Adhesive Resin

(Fig.4)


Zinc Phosphate
Glass Ionomer
Adhesive Resin
SEM
Zr
Z
Zn
C
Si
100μm

(Fig.5)