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Effects of plasma surface treatment on the bond strength of zirconia with an adhesive resin luting agent

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The purpose of this study was to evaluate the effect of the atmospheric pressure plasma treatment as a surface treatment method on the contact angle and shear bond strength (SBS) of zirconia ceramics and the failure mode between the self-adhesive resin luting agent and zirconia. The zirconia specimens were divided into eight groups based on the surface treatment method: alumina blasting, air plasma, argon plasma (AP), Katana cleaner, ozonated water, ozonated water+AP, Katana cleaner+AP, and tap water+AP. The contact angles, SBS, and fracture modes were tested. AP treatment significantly reduced the contact angle ($p<0.0001$). The combination of AP and other cleaning methods showed a higher bond strength and more mixed fractures. Our findings indicate that using atmospheric pressure plasma with argon gas, combined with other cleaning methods, results in a stronger bond than when using alumina blasting alone.

Keywords: Adhesive dentistry, Argon, Contact angle, Ozone, Y-TZP

INTRODUCTION

Zirconia is a ceramic commonly used in dentistry[1)]. Dental zirconia has been used in dentistry for more than 20 years and is now an indispensable material in clinical practice because of its high strength, toughness, and biocompatibility[1,2)]. At the time of its introduction into clinical dentistry, zirconia was a tetragonal polycrystalline material (TZP: tetragonal zirconia polycrystalline, first generation, strengths 1,000–1,500 MPa in flexure)[1)] with yttria content of 3 mol% (3Y), and was used as a framework. Since first generation zirconia is impermeable, it was layered with a feldspar-based porcelain material and applied clinically as porcelain-layered zirconia crowns, which are still widely used for anterior and molar teeth[3)]. The clinical outcomes of porcelain-layered zirconia crowns have shown that their survival rate is almost the same as that of porcelain-fused metal crowns[4)]. Subsequently, zirconia materials were improved, and translucent zirconia (3Y-TZP, second generation, strengths 900–1,300 MPa in flexure)[1)] with reduced alumina content and improved translucency was introduced[5)]. Second generation zirconia made it possible to use a single zirconia material for clinical applications in the molar region; however, it was not esthetic enough to be applied to the anterior region. The latest dental zirconia has entered the third generation, and a highly translucent partially stabilized zirconia (PSZ: third generation, strengths 400–1,000 MPa in flexure)[1)] with an increased yttria content of 4 or 5 mol% (4Y, 5Y) has been introduced.

The crystalline phase is a mixture of tetragonal and cubic crystals. Although the translucency is high and the strength is approximately half that of conventional 3Y-TZP (first generation), it can be clinically applied to the anterior teeth and molars as a monolithic zirconia crown with improved esthetic properties. Furthermore, with the development of shade-gradient zirconia discs, the gradation of zirconia crowns is now similar to that of natural teeth[6)]. The long-term clinical success of all-ceramic restorations has been reported to be closely related to the successful bonding of the restoration to a resin luting agent[7)]. Zirconia does not respond to common chemical modifications such as hydrofluoric acid (HF) etching and silanization because of its acid resistance and low silica content[8)]. Previous studies on bonding between zirconia and adhesive resin luting agents have emphasized the importance of surface preparation[7,9)]. The most common zirconia adhesion protocol consists of surface and chemical adhesion. Alumina blasting, Locatec treatment, and primer treatment with 10-methacryloyloxydecyl dihydrogen phosphate (MDP) monomers are considered effective methods[10,11)]. The goal of pretreatment is to increase surface energy and wettability by generating roughness[12)]. However, it has been reported that the alumina particle size, insufficient pressure, and inadequate treatment time during blast treatment can lead to crack formation and phase changes, which can compromise the success of zirconia restorations[13-15)]. In clinical practice, the inner surface of zirconia should be thoroughly cleaned after the intraoral fitting of a zirconia dental prosthesis to

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remove adhesive inhibitors. It has been reported that when zirconia is treated with water or phosphoric acid to remove contamination, the chemical reaction necessary for bonding occurs first, and conversely, the bond strength decreases[16,17]; therefore, care must be taken when cleaning zirconia surface.

In recent years, plasma technology has been introduced to enhance the biomaterial performance[18]. Plasma is a partially ionized gas known as the fourth state of matter[18-20]. There are two types of plasma based on how they are generated: thermal and non-thermal[21]. Thermal plasmas are generated at high pressures and temperatures and exhibit local thermodynamic equilibrium. In contrast, nonthermal plasmas are generated at close to room temperatures at atmospheric or reduced pressure (vacuum) and are characterized by differences in the energy levels of electrons and heavy particles[22,23]. Atmospheric pressure plasma is used for surface treatments such as cleaning, etching, and depositing thin surface layers[24], and has been widely applied as a new method to inactivate bacteria on the surfaces of fresh foods.

Medical devices are recognized as an economical and effective surface modification method[25,26]. The introduction of atmospheric pressure plasma has made it possible to apply plasma to the dental field, and application examples such as the inactivation of oral pathogens[19,27], teeth whitening[28], modification of implant surfaces to promote osteointegration and soft tissue healing[19,29,30], and increased interfacial adhesion of dentin and adhesive materials[22,31] have been reported. Additionally, plasma has been used to bond polymethyl methacrylate-based (PMMA)-based resin blocks[32] and adhesive resin luting agents to dentin[24], titanium, and zirconia[33,34]. Plasma techniques are used to remove contaminants and improve adhesion[35]; the cleaning mechanism depends on the gas used. Oxygen plasma promotes the formation of active reactive species and initiates surface changes, even in inert materials such as dense crystalline ceramics[18]. Argon plasma (AP) is commonly used in surface treatment technology owing to its low excitation energy, high cleaning effect, and reduced surface oxidation due to physical ablation caused by ion bombardment. Additionally, the fluorinated gas-phase treatment of zirconia with sulfur hexafluoride (SF6) has also been shown to be effective in increasing the surface reactivity[36]. The application of AP increases the polar component of the substrate, resulting in a more hydrophilic surface with improved wettability[37]. Plasma not only increases the surface energy of many materials with high efficiency, but is also used to reduce bacteria and odors, and is a promising method for cleaning, making them hydrophilic, and improving adhesion[25,26,38]. However, it has been reported that several factors such as the gas type and exposure time affect the bonding strength during plasma treatment[38]; however, no consensus has yet been reached.

The purpose of this study was to evaluate the effect of the atmospheric pressure plasma surface treatment method on the contact angle and shear bond strength (SBS) of zirconia ceramics, and failure mode between the self-adhesive resin luting agent and zirconia. The hypothesis was that atmospheric pressure plasma treatment, when used as a surface treatment, would improve the bond strength of zirconia ceramics.

MATERIALS AND METHODS

Preparation of specimens

Table 1 lists the materials used in the experiments. Zirconia containing 3 mol% yttria (Prettau Zirconia, Zirkonzahn, Gais, Italy) was cut without water injection using a silicon carbide cut-off wheel (10S20, Struers, Copenhagen, Denmark) and a precision cut-off machine (Secotom-50, Struers). Zirconia specimens were sintered in a standard high-temperature furnace (Zirkonofen Sintering Furnace, Zirkonzahn). One hundred and ninety-two zirconia plates were fabricated with a side of 10 mm and a thickness of 2 mm. The sintered zirconia plates were embedded in acrylic resin blocks (Vertex Self-Curing, Vertex Dental, Soesterberg, The Netherlands) to ensure that one surface of the disc

Table 1 Materials used

Materials	Trade name	Manufacturer	Composition	Lot No.
Zirconia ceramics	Prettau zirconia	Zirkonzahn	3 mol% yttria containig tetragonal zirconia polycrystal	ZA70029L
Alumina oxide particle	Korox 50	Bego	99.6 % aluminum oxide	14957410713
Cleaner	Katana cleaner	Kuraray Noritake Dental	10-methacryloyloxydecyl dihydrogen phosphate, purified water, triethanolamine thickeners, colorants, stabilizers	CL0025
Adhesive resin luting agent	G-cem one EM	GC	A paste: fluoroaluminosilicate glass, methacrylic acid ester, polymerization initiator B paste: silica filler, methacrylate ester, phosphate ester monomer, polymerization initiator	2210131

remained uncovered during the bonding procedures. All specimens were wet-ground with #500 grit silicon carbide abrasive (SiC Waterproof, Silicon Carbide Grinding Paper, Struers), followed by #1200 grit silicon carbide abrasive. All specimen surfaces were treated with airborne particle abrasion with 50 µm grain sized aluminum oxide particles (Korox 50, BEGO, Lincoln, RI, USA) using Cojet Prep (3M ESPE, Maplewood, MN, USA) at a pressure of 0.25 MPa from approximately 10 mm of distance for 10 s. After airborne-particle abrasion, the specimens were ultrasonically cleaned with distilled water for 10 min and air dried. They were randomly assigned to one of eight groups for the surface treatment methods.

Surface treatment methods

The bonding surfaces of each zirconia group were subjected to surface treatment (Fig. 1).

- Group CO: Control, resin luting agent applied to zirconia with alumina blasting.
- Group P: Treated with air plasma at a distance of 5 mm from the specimen for 30 s using a low-temperature atmospheric pressure plasma device (Piezobrush PZ2, Relyon Plasma, Regensburg, Germany).
- Group AP: Treated with argon plasma at a distance of 5 mm from the specimen for 30 s using a low-temperature atmospheric pressure plasma device (Piezobrush PZ2).
- Group K: The specimens were scrubbed with a microbrush using a Katana cleaner (Kuraray Noritake Dental, Japan) for 10 s, rinsed with tap water, and dried with an air syringe, according to the manufacturer's recommendations.
- Group OZ: The specimens were treated with 1.0 ppm ozonated water (Hydzonator, Hydzone, Helsinki, Finland) for 5 min and dried using an air syringe.
- Group OZAP: The OZ and AP conditions were combined.
- Group APK: The AP and K conditions were combined.
- Group WAP: A combination of tap water rinsing, drying with an air syringe, and AP.

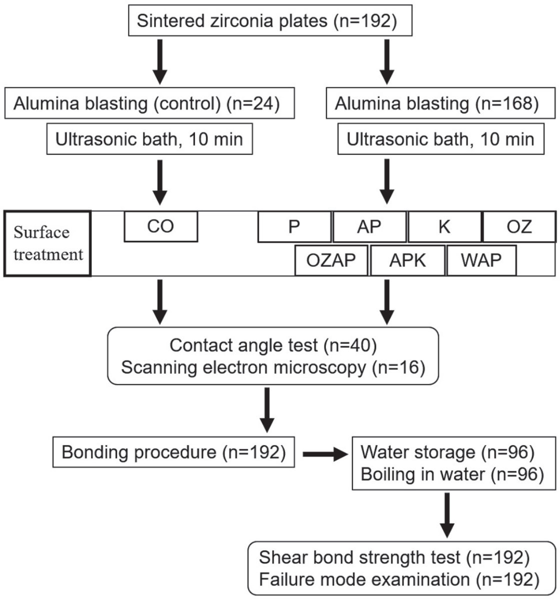

Fig. 1 Flow chart of the experimental setup.
CO: control, P: plasma with air, AP: plasma with argon gas, K: Katana cleaner, OZ: ozonated water, OZAP: OZ+AP, APK: AP+K, WAP: tap water+AP.

Contact angle measurement

Zirconia surface activation was tested by contact angle evaluation before the SBS test. Five specimens from each group were analyzed for hydrophilicity according to the zirconia surface treatment methods. Three microliters of distilled water was dropped into the center of each specimen using a microsyringe with a video contact angle goniometer (Attension Theta, Biolin Scientific, Gothenburg, Sweden), and the contact angle was measured after 10 s. Each droplet image was immediately collected at ×300 magnification, and the angles of the two opposite sides were measured and calculated as one value. The average of three measurements per specimen was used.

Bonding procedure

A translucent polyethylene mold with an inner diameter of 3.6 mm and a height of 5 mm was centered on the zirconia surface of each specimen. A self-adhesive resin luting agent paste (G-cem One EM A2, GC, Tokyo, Japan) was mixed by hand for 10 s. Subsequently, the mixed resin luting agent was placed in a syringe and filled into the mold. Excess cement was removed with a microbrush, and the samples were exposed to light perpendicular to the adhesive interface from the four directions for 20 s to cure using a light irradiator (Elipar DeepCure LED Curing Light, 3M ESPE, St. Paul, MN, USA). The specimens were stored at room temperature for 24 h after bonding.

Aging methods

One of the aging methods was water storage (WS). For this method, 12 specimens from each group were placed in plastic containers, and distilled water was added. Then, the containers were placed into an incubator at 37°C for 24 h. The other aging procedure was boiling in water (BW). For this method, distilled water was placed in glass bottles, and their caps were tightened. Then, the bottles were placed into a safety drying chamber (FDL 115, Binder, Tuttlingen, Germany) at 100°C for 24 h[39,40].

SBS testing

The specimens were mounted on the jig of a universal testing machine (LR30K Plus, Lloyd Instruments, West Sussex, UK). The cross-head speed of continuous loading was 1.0 mm/min until fracture or debonding occurred and the load deflection curve was recorded with Nexygen

4.0 software (Lloyd Instrument).

Failure mode observation and surface analysis using scanning electron microscopy (SEM)

Specimens were sputter-coated with gold ((Balzers SCD 050 sputter coater; B.U.A., Fürstentum, Germany) and evaluated under SEM (JSM-5500LV, JEOL, Tokyo, Japan) at 25kV and 2,000× magnification, before bonding procedure. After SBS testing, the failure mode was evaluated using stereo zoom microscopy (Leica Wild M3z, Leica, Heerbrugg, Switzerland) at ×20 magnification and the mode of failure was expected to be of three different types: cohesive failure that occurs within the resin luting agent, adhesive failure that occurs between the resin luting agent and zirconia material interface, and mixed failure or a combination of both failure modes[41].

Statistical analysis

Bartlett's test was used for contact angle and SBS testing. When homoscedasticity was shown in the obtained measured values, Dunnett's test was performed as a comparison test with the control group after one-way analysis of variance. If homoscedasticity was not observed, the Kruskal–Wallis test was performed, and the Steel test was performed for comparison with the control group. Additionally, for the SBS test, the Shapiro-Wilk W test was primarily used to determine whether the distribution was normal, and *t*-tests were performed to compare the aging methods within the same group ($p<0.05$). All statistical analyses were performed using JMP Pro (JMP Pro 17.0.0, SAS Institute, Cary, NC, USA).

RESULTS

The contact angle measurements are shown in Fig. 2. The contact angle of the water droplets in group APK was the highest at 90.4±3.7°. Groups K and APK using Katana cleaner showed higher contact angles than the other groups. In contrast, groups P and OZ had lower contact angles than the control group (55.9±7.9°). Furthermore, it was not possible to measure this in the AP, OZAP, and WAP groups. Therefore, these three groups were excluded from the statistical analyses. Dunnett's test results showed significant differences from the control group under all conditions ($p<0.0001$).

The SEM images at 2,000× magnification showing

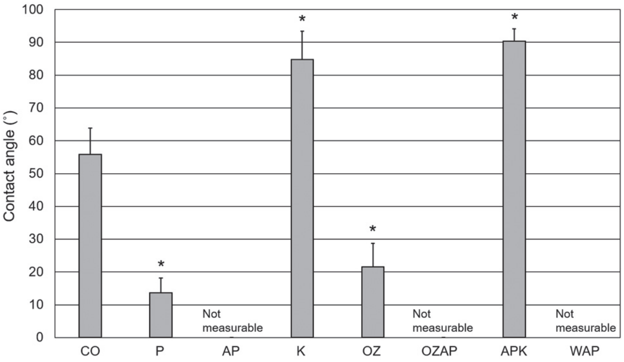

Fig. 2 Contact angle results after surface treatment of zirconia.
* indicates a significant difference compared to the control group ($p<0.05$).

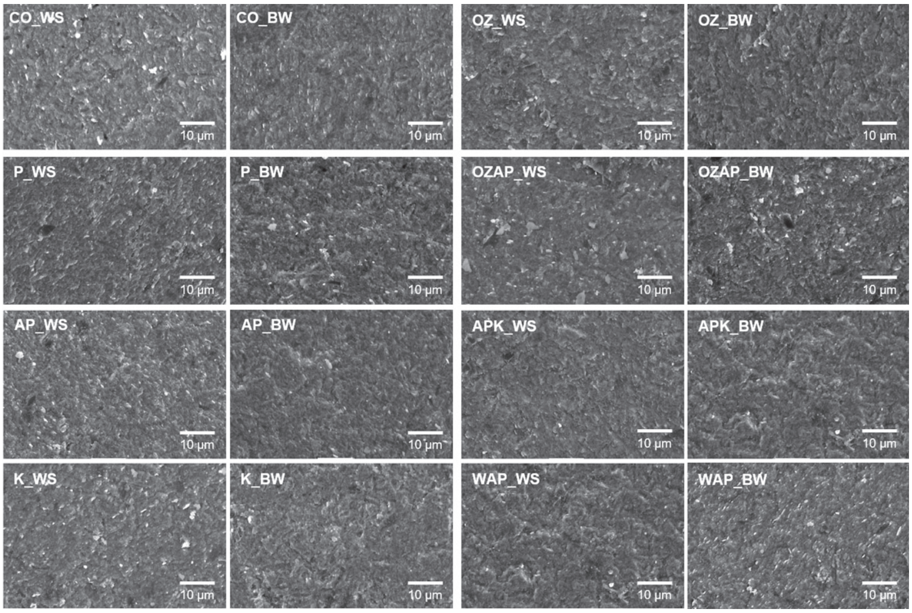

Fig. 3 SEM images at 2,000× magnification after surface treatment of zirconia.
CO: control, P: plasma with air, AP: plasma with argon gas, K: Katana cleaner, OZ: ozonated water, OZAP: OZ+AP, APK: AP+K, WAP: tap water+AP, WS: water storage, BW: boiling in water.

the microstructures of the zirconia specimens after surface treatments (Fig. 3). SEM images showed no difference in surface structure between the different surface treatment and aging methods.

In the SBS test, group K of WS had the highest value (24.4±1.3 MPa), and group OZ of WS had the lowest (10.1±0.7 MPa). The SBS tended to be higher in WS than in BW in more than half of the groups. However, in groups OZ, OZAP, and WPG, WS tended to have a higher bond strength than BW (Fig. 4). In the Steel test, a significant difference in the WS was observed between the OZ (p=0.0059) and control groups. In contrast, significant differences in BW were observed between the groups OZAP (p=0.0059), APK (p=0.0085), and WAP (p=0.0059) and the control group. The Shapiro-Wilk W test showed a p-value greater than 0.05. Regarding the aging method for the same surface treatment, significant differences were observed among CO (p=0.0008), K (p<0.0001), OZ (p<0.0001), and WAP (p=0.0217).

The SEM images show representative examples of failure modes for each test condition (Fig. 5). Failure mode testing revealed adhesion or mixed failures;

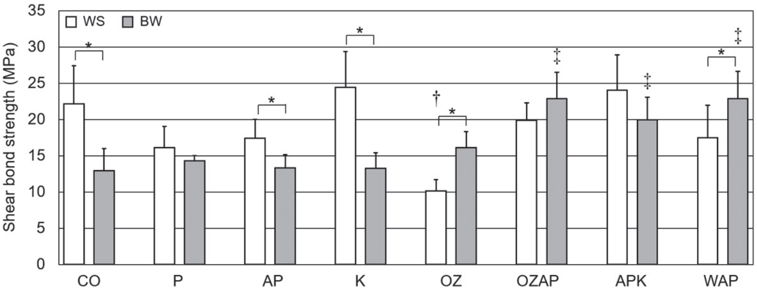

Fig. 4 SBS test results.
CO: control, P: plasma with air, AP: plasma with argon gas, K: Katana cleaner, OZ: ozonated water, OZAP: OZ+AP, APK: AP+K, WAP: tap water+AP, WS: water storage, BW: boiling in water. * indicates significant differences between the aging methods within the same surface treatment (p<0.05). † indicates a significant difference in WS from the control group (p<0.05). ‡ indicates a significant difference in BW from the control group (p<0.05).

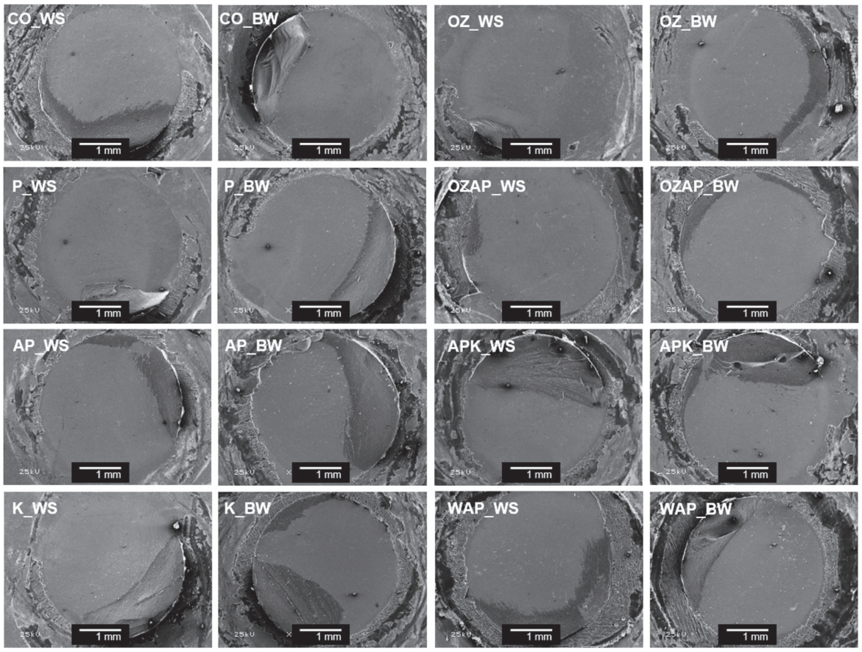

Fig. 5 SEM images at 25× magnification after SBS test.
CO: control, P: plasma with air, AP: plasma with argon gas, K: Katana cleaner, OZ: ozonated water, OZAP: OZ+AP, APK: AP+K, WAP: tap water+AP, WS: water storage, BW: boiling in water.

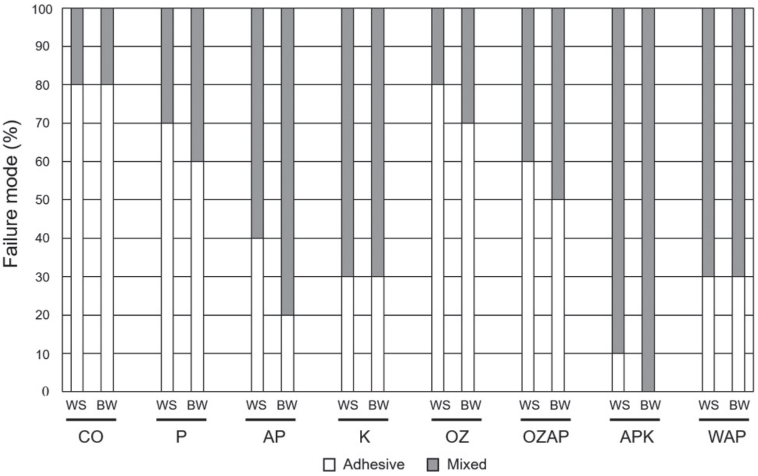

Fig. 6 Failure mode of the zirconia specimens after SBS test (%).
CO: control, P: plasma with air, AP: plasma with argon gas, K: Katana cleaner, OZ: ozonated water, OZAP: OZ+AP, APK: AP+K, WAP: tap water+AP.

however, cohesive failure was not observed. In the APK group, mixed failure was observed in most specimens. Mixed fractures were most common in the AP, K, and APK groups (AP >60%; K >70%; APK >90%). However, adhesive failure was more common in the OZ and control groups (>70%) (Fig. 6).

DISCUSSION

In this study, the effect of atmospheric pressure plasma treatment on the bond strength of MDP-containing self-adhesive resin luting agents with zirconia ceramics was evaluated. atmospheric pressure plasma treatment did not affect the bond strength of the adhesive resin luting agent and zirconia at 37° WS; however, it improved the bond strength at BW; thus, the null hypothesis was partially rejected.

In vitro studies and clinical durability evaluations of various adhesion protocols of 3Y-TZP to enamel, dentin, composite resin, titanium, and zirconia have been reported[42)], and 3Y-TZP was used in this study to discuss them with the literature.

In dentistry, AP treatment has shown promising results in enhancing resin adhesion to zirconia ceramics[37,43)]. The introduction of plasma in surface treatments improves adhesion by increasing the surface energy of the material without increasing its roughness or causing less damage than alumina particles[13,37)]. In this study, aging methods such as WS and boiling in distilled water were adopted as common aging methods mentioned in the literature[39,40)]. The low-temperature atmospheric pressure plasma device used here was designed as a compact handheld plasma device for use in laboratories and in small series assemblies. With a maximum power consumption of 18 W, the piezoelectric direct discharge technology was used to generate active plasma at a low temperature. Among the various plasma treatments, atmospheric pressure plasma is more feasible in dental laboratories and chairside work to improve adhesive resin luting agents for zirconia bonding because it does not require a vacuum chamber or equipment[37,44,45)].

Measuring the contact angle is a common method for calculating surface energies indicative of surface wettability[46)]. In this study, the contact angles could not be measured for the groups using AP other than the APK group, which indicated that AP treatment increased the hydrophilicity of the zirconia surface. The use of AP increased the hydrophilicity of the zirconia surface, which is consistent with the results of other studies[37,47)]. The application of AP to the zirconia surface removes organic residues, promotes surface restructuring, reduces carbon, and increases oxygen[37,47)]. It has been suggested that the increased hydrophilicity of the zirconia surface is associated with an increase in the number of oxygen atoms on the surface, thereby making it more polar. This has reportedly improved wetting, reduced the surface contact angle, and promoted resin adhesion[37,48)]. Atmospheric pressure plasma treatment has been reported to increase the oxygen content and decrease the carbon content of the zirconia surface[38)], which may increase the surface energy and improve surface wettability.

The SBS values for all groups were above 10 MPa. Initial SBS test data give a first impression of the bonding, but ultimately only data after aging or long-term storage may be combined to estimate the durability of the bonding. In this regard, SBS values above 10 MPa have been reported to be acceptable[49)]. Thus, it was considered a threshold for good bond strength between the resin luting agent and restoration. The group that received AP treatment and other surface treatment methods achieved higher SBS values even after aging of BW, and showed SBS values equal to or better than those of the control group. This result is consistent with previous studies that used AP treatment[37,38]. Furthermore, the decrease in the SBS value after heat aging is similar to that reported in other studies[47]. In this study, the bond strength between the control and AP groups was almost the same; therefore, AP treatment was not considered a substitute for alumina blasting. This differs from the report by Valverde *et al.*, who observed a significant increase in the bond strength to zirconia surfaces even when AP was applied alone[37]. It has been reported that plasma treatment with helium improves the bond strength of zirconia and adhesive resin luting agents as effectively as alumina airborne-particle abrasion[44]. Ye *et al.* reported that helium plasma treatment improves the surface wettability of highly transparent zirconia without changing the surface morphology or roughness, and ultimately significantly improves the adhesive strength and durability of the resin luting agent. Furthermore, it has been reported that the combination of atmospheric pressure plasma treatment and MDP-containing primers further improves adhesion, especially after artificial aging, resulting in a strong and durable bond between the resin luting agent and the highly transparent zirconia[20]. It has been reported that oxygen plasma treatment may improve the initial bond strength when applied with alumina blasting or silane-MDP mixed primers. However, the SBS values decreased significantly after thermal cycling; thus, oxygen plasma is not recommended for use with MDP-based adhesives or universal adhesives[45]. It has been concluded that plasma treatment with oxygen is not a substitute for airborne-particle abrasion of zirconia bonds[50,51]. Furthermore, it has been reported that the SBS of Ce-TZP/Al_2O_3 and MDP-containing resin binders can be significantly improved by combining airborne-particle abrasion and glow discharge plasma treatment[46]. However, under the AP treatment conditions used in this study, no improvement in the SBS between zirconia and the MDP-containing adhesive resin luting agent was expected. Significant differences were observed in the BW of the OZAP, APK, and WAP groups combined with AP compared with the control group, and the SBS was over 20 MPa, indicating that combinations with other cleaning methods were effective.

In the fracture mode, the specimens treated in groups K and AP exhibited a high mixed fracture rate. Adhesive failure is thought to increase when the SBS is low. The failure modes were different from those in a previous report[38], in which no adhesive failures were observed; however, they were similar to previous reports because there were many mixed failures[37,38,47].

Ozone (O_3) is a strong oxidizing and effective antimicrobial agent. Therefore, it has been used in medicine for many years, and more recently, in dentistry[52]. Owing to its bactericidal, fungicidal, and virucidal properties, ozone is widely used in several fields of dentistry, including stomatology, endodontics, periodontology, surgery, and treatment of carious lesions[53]. In bond strength tests, the SBS between enamel, brackets, and composite resins for filling has been reported using ozone gas[54-56]. However, in this study, ozonated water was used to clean the surface of zirconia. Ozonated water is a safe aqueous solution that can be produced without the use of chemical components and has no residual properties. The strong oxidizing power of ozone (approximately seven times that of chlorine) can decompose organic stains. Additionally, tap water was added as a condition for the presence of moisture. In the OZ, OZAP, and WAP groups, in which moisture was present during surface treatment, the SBS tended to be greater in BW than in WS. If water is present on the zirconia surface, there may be some reaction with MDP. Therefore, it will be necessary to verify this in the future. The AP treatment applied in this study significantly enhanced the surface wettability by cleaning the organic impurities and promoting the physical adsorption of water. It has been reported that cleaning with tap water reduces bond strength[16,17]; however, in this study, it was considered effective in combination with AP treatment.

This study has some limitations. Only one type of zirconia was used in this study, and its composition and properties have been reported to depend on the product brand[57]. The polarity fraction differs depending on the zirconia product[48], and the presence of many nonpolar groups on the zirconia surface is due to the low hydrophilicity of zirconia ceramics and the low surface energy[37]. Therefore, the effects of atmospheric pressure plasma on zirconia manufactured by different manufacturers need to be further evaluated. In addition, further research is required to optimize the application conditions of atmospheric pressure plasma and evaluate its adhesion-promoting effect. Long-term water storage is necessary to distinguish clinically durable zirconia bonding systems from non-durable zirconia bonding systems[58].

Atmospheric pressure plasma is a promising technology for increasing the bond strength of difficult-to-bond materials, and further research should be conducted on the influence of atmospheric pressure plasma on the long-term durability of bonds to zirconia.

CONCLUSION

Within the limitations of this study, atmospheric pressure plasma with argon gas improved the wettability of the zirconia surface. The combination of AP treatment and other cleaning methods resulted in a higher bond strength than alumina blasting alone. Mixed fractures occurred more frequently in the plasma-treated group to which plasma treatment was applied. Overall, atmospheric pressure plasma is a promising technology for increasing the bond strength of difficult-to-bond materials, and further research should be conducted on the influence of atmospheric pressure plasma on the long-term durability of bonds to zirconia.

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REFERENCES

1) Zhang Y, Lawn BR. Novel zirconia materials in dentistry. J Dent Res 2018; 97: 140-147.
2) Blatz MB, Chiche G, Bahat O, Roblee R, Coachman C, Heymann HO. Evolution of aesthetic dentistry. J Dent Res 2019; 98: 1294-1304.
3) Miura S, Kasahara S, Yamauchi S, Okuyama Y, Izumida A, Aida J, *et al*. Clinical evaluation of zirconia-based all-ceramic single crowns: An up to 12-year retrospective cohort study. Clin Oral Investig 2018; 22: 697-706.
4) Sailer I, Makarov NA, Thoma DS, Zwahlen M, Pjetursson BE. All-ceramic or metal-ceramic tooth supported fixed dental prostheses (FPDs)? A systematic review of the survival and complication rates. Part I: single crowns (SCs). Dent Mater 2015; 31: 603-623.
5) Miura S, Yamauchi S, Kasahara S, Katsuda Y, Fujisawa M, Egusa H. Clinical evaluation of monolithic zirconia crowns: A failure analysis of clinically obtained cases from a 3.5-year study. J Prosthodont Res 2021; 65: 148-154.
6) Miura S, Tsukada S, Fujita T, Isogai T, Teshigawara D, Saito-Murakami K, *et al*. Effect of abutment tooth and luting agent colors on final color of high-translucent zirconia crowns. J Prosthodont Res 2022; 66: 243-249.
7) Blatz MB, Sadan A, Kern M. Resin-ceramic bonding: A review of the literature. J Prosthet Dent 2003; 89: 268-274.
8) Dérand P, Derand T. Bond strength of luting cements to zirconium oxide ceramics. Int J Prosthodont 2000; 13: 131-135.
9) Thompson JY, Stoner BR, Piascik JR, Smith R. Adhesion/cementation to zirconia and other non-silicate ceramics: Where are we now? Dent Mater 2011; 27: 71-82.
10) Grasel R, Santos MJ, Chagas Rěgo HM, Rippe MP, Valandro LF. Effect of resin luting systems and alumina particle air abrasion on bond strength to zirconia. Oper Dent 2018; 43: 282-290.
11) Comino-Garayoa R, Peláez J, Tobar C, Rodriguez V, Suárez MJ. Adhesion to zirconia: A systematic review of surface pretreatments and resin cements. Materials 2021; 14: 2751.
12) Russo DS, Cinelli F, Sarti C, Giachetti L. Adhesion to zirconia: A systematic review of current conditioning methods and bonding materials. Dent J 2019; 7: 74.
13) Hallmann L, Ulmer P, Wille S, Polonski O, Köbel S, Trottenberg T, *et al*. Effect of surface treatments on the properties and morphological change of dental zirconia. J Prosthet Dent 2016; 115: 341-349.
14) Kwon SM, Min BK, Kim YK, Kwon TY. Influence of sandblasting particle size and pressure on resin bonding durability to zirconia: A residual stress study. Materials 2020; 13: 5629.
15) Kim HK, Ahn B. Effect of Al_2O_3 sandblasting particles size on the surface topography and residual compressive stresses of three different dental zirconia grades. Materials 2021; 14: 610.
16) Yang B, Wolfart S, Scharnberg K, Ludwig K, Adelung R, Kern M. Influence of contamination on zirconia ceramic bonding. J Dent Res 2007; 86: 749-753.
17) Ishii R, Tsujimoto A, Takamizawa T, Tsubota K, Suzuki T, Shimamura Y, *et al*. Influence of surface treatment of contaminated zirconia on surface free energy and resin cement bonding. Dent Mater J 2015; 34: 91-97.
18) Chu PK, Chen JY, Wang LP, Huang N. Plasma-surface modification of biomaterials. Mater Sci Eng R Rep 2002; 36: 143-206.
19) Yang Y, Zheng M, Yang Y, Li J, Su YF, Li HP, *et al*. Inhibition of bacterial growth on zirconia abutment with a helium cold atmospheric plasma jet treatment. Clin Oral Investig 2020; 24: 1465-1477.
20) Ye XY, Lai YT, Song WP, Hu Y. Effects of cold atmospheric plasma treatment on resin bonding to high translucency zirconia ceramic. Dent Mater J 2022; 41: 896-904.
21) Conrads H, Schmidt M. Plasma generation and plasma sources. Plasma Sourc Sci Tech 2000; 9: 441-454.
22) Stancampiano A, Forgione D, Simoncelli E, Laurita R, Tonini R, Gherardi M, *et al*. The effect of cold atmospheric plasma (CAP) treatment at the adhesive-root dentin interface. J Adhes Dent 2019; 21: 229-237.
23) Altuntas M, Colgecen O, Ercan UK, Cukur E. Nonthermal plasma treatment can eliminate sandblasting procedure for zirconia-resin cement bonding. Int J Prosthodont 2022; 35: 752-760.
24) Kim JH, Han GJ, Kim CK, Oh KH, Chung SN, Chun BH, *et al*. Promotion of adhesive penetration and resin bond strength to dentin using non-thermal APP. Eur J Oral Sci 2016; 124: 89-95.
25) Takamatsu T, Uehara K, Sasaki Y, Hidekazu M, Matsumura Y, Iwasawa A, *et al*. Microbial inactivation in the liquid phase induced by multigas plasma jet. PLOS ONE 2015; 10: e0135546.
26) Namura Y, Uchida Y, Sato R, Shimizu N, Motoyoshi M, Tsutsumi Y, *et al*. Changes in surface properties of dental alloys with atmospheric plasma irradiation. Dent Mater J 2020; 39: 375-380.
27) Hui WL, Ipe D, Perrotti V, Piattelli A, Fang Z, Ostrikov K, *et al*. Novel technique using cold atmospheric plasma coupled with air-polishing for the treatment of titanium discs grown with biofilm: An in-vitro study. Dent Mater 2021; 37: 359-369.
28) Pavelić B, Švarc MZ, Šegović S, Bago I. Cold atmospheric plasma for bleaching endodontically treated tooth: A new clinical approach. Quintessence Int 2020; 51: 364-371.
29) Giro G, Tovar N, Witek L, Marin C, Silva NR, Bonfante EA, *et al*. Osseointegration assessment of chairside argon-based nonthermal plasma-treated Ca-P coated dental implants. J Biomed Mater Res A 2013; 101: 98-103.
30) Zheng M, Yang Y, Liu Xiao-Qiang, Liu Ming-Yue, Zhang Xiao-Fei, Wang X, *et al*. Enhanced biological behavior of in vitro human gingival fibroblasts on cold plasma treated zirconia. PLOS ONE 2015; 13: e0140278.
31) Zhu Xiao-Ming, Zhou Jian-Feng, Guo H, Zhang Xiao-Fei, Liu Xiao-Qiang, Li He-Ping. Effects of a modified cold atmospheric plasma jet treatment on resin-dentin bonding. Dent Mater J 2018; 37: 798-804.
32) Liebermann A, Keul C, Bähr N, Edelhoff D, Eichberger M, Roos M, *et al*. Impact of plasma treatment of PMMA-based CAD/CAM blanks on surface properties as well as on adhesion to self-adhesive resin composite cements. Dent Mater 2013; 29: 935-944.
33) Silva NR, Coelho PG, Valverde GB, Becker K, Ihrke R, Quade A, *et al*. Surface characterization of Ti and Y-TZP following non-thermal plasma exposure. J Biomed Mater Res B Appl Biomater 2011; 99: 199-206.
34) Kobune K, Miura T, Sato T, Yotsuya M, Yoshinari M. Influence of plasma and ultraviolet treatment of zirconia on initial attachment of human oral keratinocytes: Expressions of laminin γ2 and integrin β4. Dent Mater J 2014; 33: 696-704.
35) Aronsson BO, Lausmaa J, Kasemo B. Glow discharge plasma treatment for surface cleaning and modification of metallic biomaterials. J Biomed Mater Res 1997: 35; 49-73.
36) Piascik JR, Swift EJ, Braswell K, Stoner BR. Surface fluorination of zirconia: Adhesive bond strength comparison to commercial primers Dent Mater 2012; 28: 604-608.
37) Valverde GB, Coelho PG, Janal MN, Lorenzoni FC, Carvalho RM, Thompson VP, *et al*. Surface characterization and

bonding of Y-TZP following non-thermal plasma treatment. J Dent 2013; 41: 51-59.

38) Sevilla P, Gseibat M, Peláez J, Suárez M, López-Suárez C. Effect of surface treatments with low-pressure plasma on the adhesion of zirconia. Materials 2023; 16: 6055.

39) Brendeke J, Özcan M. Effect of physicochemical aging conditions on the composite-composite repair bond strength. J Adhes Dent 2007; 9: 399-406.

40) Egilmez F, Ergun G, Cekic-Nagas I, Vallittu P, Lassila LV. Does artificial aging affect mechanical properties of CAD/CAM composite materials. J Prosthodont Res 2018; 62: 65-74.

41) Pisani-Proenca J, Erhardt MCG, Valandro LF, Gutierrez-Aceves G, Bolanos-Carmona MV, Castillo-Salmeron RD. Influence of ceramic surface conditioning and resin cements on microtensile bond strength to a glass ceramic. J Prosthet Dent 2006; 96: 412-417.

42) Alammar A, Blatz MB. The resin bond to high-translucent zirconia —A systematic review. J Esthet Restor Dent 2022; 34: 117-135.

43) Elias AB, Simão RA, Prado M, Cesar PF, Santos GBD, da Silva EM. Effect of different times of nonthermal argon plasma treatment on the microtensile bond strength of self-adhesive resin cement to yttria-stabilized tetragonal zirconia polycrystal ceramic. J Prosthet Dent 2019; 121: 485-491.

44) Ito Y, Okawa T, Fukumoto T, Tsurumi A, Tatsuta M, Fujii T, *et al*. Influence of atmospheric pressure low-temperature plasma treatment on the SBS between zirconia and resin cement. J Prosthodont Res 2016; 60: 289-293.

45) Kang LL, Chuang SF, Li CL. Enhancing resin cement adhesion to zirconia by oxygen plasma-aided silicatization. Materials 2022; 15: 5568.

46) Egoshi T, Taira Y, Sakihara M, Kamada K, Sawase T, Murata H. Effects of abrasion and glow-discharge plasma treatment on bonding resin cement to ceria-stabilized zirconia/alumina nanocomposite. Dent Mater J 2019; 38: 437-443.

47) Kim DS, Ahn JJ, Bae EB, Kim GC, Jeong CM, Huh JB, *et al*. Influence of non-thermal atmospheric pressure plasma treatment on shear bond strength between Y-TZP and delf-adhesive resin cement. Materials 2019; 12: 3321.

48) Negreiros WM, de Souza VTFS, Lopes BB, Giannini M. Adhesion of resin cement to zirconia using argon plasma and primer. Int J Prosthodont 2021; 34: 796-800.

49) Rosentritt M, Preis V, Behr M, Sereno N, Kolbeck C. Shear bond strength between veneering composite and PEEK after different surface modifications. Clin Oral Investig 2015; 19: 739-744.

50) Liu YC, Hsieh JP, Chen YC, Kang LL, Hwang CS, Chuang SF. Promoting porcelain-zirconia bonding using different atmospheric pressure gas plasmas. Dent Mater 2018; 34: 1188-1198.

51) Lümkermann N, Eichberger M, Stawarczyk B. Different surface modifications combined with universal adhesives: The impact on the bonding properties of zirconia to composite resin cement. Clin Oral Investig 2019; 23: 3941-3950.

52) Holmes J. Clinical reversal of root caries using ozone, double-blind, randomized, controlled 18-month trial. Gerodontology 2003; 20: 106-114.

53) El Meligy OA, Elemam NM, Talaat IM. Ozone therapy in medicine and dentistry: A review of the literature. Dent J 2023; 11: 187.

54) Magni E, Ferrari M, Papacchini F, Hickel R, Ilie N. Influence of ozone on the composite-to-composite bond. Clin Oral Investig 2011; 15: 249-256.

55) Rodrigues P, Souza JB, Soares CJ, Lopes LG, Estrela C. Effect of ozone application on the resin-dentin microtensile bond strength. Oper Dent 2011; 36: 537-544.

56) Cossellu G, Lanteri V, Butera A, Sarcina M, Farronato G. Effects of six different preventive treatments on the shear bond strength of orthodontic brackets: In vitro study. Acta Biomater Odontol Scand 2015; 1: 13-17.

57) Sulaiman TA, Abdulmajeed AA, Donovan TE, Ritter AV, Vallittu PK, Närhi TO, *et al*. Optical properties and light irradiance of monolithic zirconia at variable thicknesses. Dent Mater 2015; 31: 1180-1187.

58) Wegner SM, Kern M. Long-term resin bond strength to zirconia ceramic. J Adhesive Dent 2000; 2: 139-147.