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ORIGINAL ARTICLE

Effect of non-thermal argon plasma on the shear strength of adhesive systems

Isabella de Almeida Guimarães Passos[1], Juliana das Neves Marques[1], João Victor Frazão Câmara[2]*, Renata Antoun Simão[3], Maíra do Prado[1] and Gisele Damiana da Silveira Pereira[1]

[1]Departamento de Clínica Odontológica, Faculdade de Odontologia, Universidade Federal do Rio de Janeiro – UFRJ, Rio de Janeiro, RJ, Brasil

[2]Clinic of Operative Dentistry, Periodontology and Preventive Dentistry, Saarland University Hospital, Homburg, Saar, Germany

[3]Departamento de Engenharia Metalúrgica e de Materiais, Centro de Tecnologia, Universidade Federal do Rio de Janeiro – UFRJ, Rio de Janeiro, RJ, Brasil

**jvfrazao92@hotmail.com*

Abstract

This study evaluated the influence of non-thermal argon plasma applied to dentin on the shear strength of two adhesive systems. Ninety tooth fragments were embedded in epoxy resin and distributed into experimental groups (n=15): G1 and G4 - adhesive systems applied according to the manufacturers' instructions; G2 and G5 - dentin treated with non-thermal argon plasma for 30 seconds before hybridization; G3 and G6 - dentin treated with non-thermal argon plasma for 30 seconds after hybridization. Cylinders were made with composite resin in the adhesive area, and the specimens were submitted to the shear strength test. Higher values were observed when applying the plasma treatment after hybridization (G1: 26.51 MPa, G2: 29.22 MPa, G3: 30.27 MPa, G4: 22.66 MPa, G5: 28.33 MPa, G6: 29.32 MPa). The treatment with non-thermal argon plasma significantly increased the shear strength values regardless of the application time.

Keywords: *argon plasma, non-thermal plasma, dentin bonding agents, dentin, shear strength.*

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1. Introduction

Several factors inherent to the physicochemical structure of adhesives and their intrinsic properties interfere with the formation of the hybrid dentin layer[1-4]. The morphological, physiological, and pathological heterogeneity of dentin, the moisture needed to maintain the expanded collagen network, its low surface energy, and the limited degree of conversion of resin monomers are among the main obstacles to achieving uniform adhesion[5]. Moreover, the hydrolytic degradation of both components of the hybrid layer - collagen matrix and composite resin - seriously compromises the long-term adhesive interface integrity and the bond strength durability, causing postoperative sensitivity, bacterial microleakage, and secondary caries[4,5].

Non-thermal plasmas are considered the fourth state of matter, comprising partially ionized gases with different concentrations of highly reactive low molecular weight particles, including electronically excited atoms, molecules, ionic species, and free radicals applied at temperatures close to body temperature, which allows using them *in vivo*[6,7]. Plasma has been used in the surface engineering industry for improving biomaterial adhesion by depositing thin films[8]. When used correctly, the non-thermal plasma modifies the physical and chemical properties of surfaces, maintaining the interior characteristics of the material[9]. Recent studies have proposed using plasma technology in Dentistry for different purposes, including dental caries treatment, sterilization, biofilm elimination, root canal disinfection, and tooth whitening, among others[6,10,11]. Furthermore, plasma has been used to improve adhesion to the dental substrate because treatments with this gas increase the contact surface area of collagen fibers and their hydrophilicity[6,10], allowing a higher interaction with the adhesive and increasing bond strength[6,9]. Plasma also induces and increases the degree of conversion of resin monomers.

The high complexity of the dentin tissue and the factors that influence bond durability require scientific evaluations of bond strength and surface treatments. Thus, this study aimed to evaluate the influence of non-thermal argon plasma applied to deep dentin, before and after hybridization, on the shear strength of two adhesive systems: a conventional three-step system and a self-etching single-step system. The following null hypotheses were tested: 1 - The composition and application technique of adhesive systems do not affect adhesive strength; 2 - The application of non-thermal argon plasma does not affect the shear strength of adhesive systems; 3 - The application of non-thermal argon

plasma before the adhesive systems does not interfere with adhesion; 4 - The application of non-thermal argon plasma on hybridized dentin does not interfere with the shear strength of adhesive systems.

2. Materials and Methods

2.1 Ethical aspects

The research project was submitted to and approved by the Research Ethics Committee of the University Hospital Clementino Fraga Filho (HUCFF) of the Federal University of Rio de Janeiro - UFRJ (RJ, Brazil), with approval number 79803517.6.0000.5257.

2.2 Materials

Two adhesive systems were used: a conventional water-based multi-bottle system (Adper Scotchbond Multi-Purpose, 3M do Brasil, Sumaré, SP, Brazil) and a single-bottle self-etching ethanol-based system (Single Bond Universal, 3M do Brasil, Sumaré, SP, Brazil). The study also used an Opallis nanohybrid restorative composite in A3 color (FGM LTDA, Joinville, SC, Brazil), Condac 37% phosphoric acid conditioner (FGM LTDA, Joinville, SC, Brazil), and non-thermal plasma from Argon gas (White Martins, Rio de Janeiro, RJ, Brazil). Table 1 describes the brands and compositions of the materials selected.

2.3 Experimental groups

The samples were divided into six experimental groups with 15 repetitions each, as follows: Group 1 – SBMPSP: after acid etching, the Adper Scotchbond Multi-Purpose adhesive system was applied to the dentin surface; Group 2 – SBMPPA: after acid etching, argon plasma was applied for 30 seconds to the dentin surface, followed by the application of Adper Scotchbond adhesive; Group 3 - SBMPPD: after acid etching, Adper Scotchbond Multi-Purpose adhesive was applied, followed by the application of argon plasma for 30 seconds; Group 4 – SBUSP: the Single Bond Universal adhesive system was applied to the dentin surface; Group 5 – SBUPA: argon plasma was applied for 30 seconds to the dentin surface, followed by the application of the Single Bond Universal adhesive system; Group 6 – SBUPD: the Single Bond Universal adhesive system was applied to the dentin surface, followed by the application of argon plasma for 30 seconds.

2.4 Preparation of dentin samples

The study used 45 human third molars impacted and freshly extracted by therapeutic indication. The teeth were collected in a private office after patients had signed the donation terms and the informed consent form.

The teeth were stored for up to 30 days in a 0.1% thymol solution (UFRJ-CCMN, Department of Biochemistry, Rio de Janeiro, RJ, Brazil) at a pH of 7 for disinfection, and a temperature of 37ºC in an oven (Quimis Scientific Apparatus, São Paulo, SP, Brazil) until starting the external surface cleaning with # 13/14 Gracey periodontal curettes (Hu-Friedy do Brasil, Rio de Janeiro, RJ, Brazil) to remove periodontal tissue residues. Subsequently, the teeth were submitted to prophylaxis with a pumice stone (SS-White, Rio de Janeiro, RJ, Brazil) and water, using Robinson brushes (KG Sorensen, Barueri, SP, Brazil) mounted in a counter-angle at low rotation (Kavo do Brazil, Joinville, SC, Brazil).

The coronal portions of the 45 upper or lower third molars were separated from their roots 1 mm below the cemento-enamel junction with a double-sided diamond disc (KG Sorensen, Barueri, SP, Brazil) at low rotation and under abundant water cooling/air. Then, the fragments obtained were sectioned in two parts mesiodistally from the occlusal surface with the same process, producing 90 tooth fragments.

The enamel surface of each fragment was sanded and fixed on a glass plate aided by adhesive tape to facilitate their inclusion in the epoxy resin (Redecenter Materials Plásticos e Acessórios LTDA, São Paulo, SP, Brazil) poured into PVC tube rings with 21 mm in internal diameter and

Table 1. Brands and composition of materials.

Material	Composition	Manufacturer	Classification
Condac 37%	37% phosphoric acid	FGM LTDA, Joinville, SC, Brazil	Acid conditioner
Adper Scotchbond Multi-Purpose	Primer: Aqueous solution of 2-hydroxyethyl methacrylate (HEMA) and a copolymer. Adhesive: Bisphenol-a-glycidyl methacrylate (Bis-GMA), 2-hydroxyethyl methacrylate (HEMA), and camphorquinone solution.	3M do Brasil, Sumaré, SP, Brazil	Conventional 3-step adhesive system
Single Bond Universal	Bisphenol-a-diglycidyl ether dimethacrylate (Bis-GMA), 2-hydroxyethyl methacrylate, water, 1,10-decanediol methacrylate phosphate, acrylic, itaconic acid copolymer, camphorquinone, N,N-dimethylbenzoin, 2-dimethyl monoethyl methacrylate, and methyl ethyl ketone.	3M do Brasil, Sumaré, SP, Brazil	Self-etching single-step adhesive system
Opallis	Monomeric matrix: Bis (GMA), Bis (EMA), UDMA, and TEGDMA. Fillers: Barium-aluminosilicate glass, silanized barium-aluminosilicate glass, and silicon dioxide nanoparticles. Photoinitiator: camphorquinone, accelerators, stabilizers, and pigments. The composite particles range from 40 nm to 3.0 microns with an average particle size of 0.5 microns, total filler content by weight from 78.5% to 79.8%, and volume from 57% to 58% of inorganic filler.	FGM LTDA, Joinville, SC, Brazil	Nanohybrid resin
Non-thermal plasma	Argon gas.	White Martins, Rio de Janeiro, RJ, Brazil	Propagation gas non-thermal plasma

10 mm in height. These tubes were also fixed on the glass plate to centralize the tooth fragments[12].

After polymerization, the epoxy resin blocks (Redecenter Materials Plásticos e Acessórios LTDA, São Paulo, SP, Brazil) were removed from the PVC cylinders and the buccal, palatal, and lingual surfaces were abraded with a #180 silicon carbide sandpaper (3M do Brasil, Sumaré, SP, Brazil) in a water-cooled rotary electric polisher (Aropol 2V, Arotec Indústria e Comércio, Cotia, SP, Brazil) to expose a flat area in the deep dentin. The control distance from the dentin surface to the pulp chamber was based on the protocol by dos Santos et al.[13]. Finishing was performed similarly with a #600 silicon carbide sandpaper. Then, the flat surfaces (3D) were washed with distilled water spraying for 15 seconds. The samples were identified so they could be distributed by drawing among the experimental groups. The specimens were stored in containers with distilled water and maintained in an oven at 37ºC while awaiting the preparation of specimens for the mechanical test.

2.5 Non-thermal plasma application

The non-thermal plasma treatment was performed with a glass reactor, which consisted of a glass tube of 5 cm in diameter and 30 cm in length, evacuated by a mechanical pump to pressures below 2 Pascals (Pa). The gas was allowed to fill the reactor up to 10 Pa of pressure[6]. The non-thermal plasma was produced inside the glass cylinder under vacuum by the action of a magnetic field induced by the current passing through an electric coil surrounding the cylinder. The dentinal surfaces were treated with argon gas at 60 watts (w) for 30 seconds[6]. At the end of the process, the radio frequency was turned off before exposing the samples to air. In groups 2 and 5, this procedure was performed before applying the adhesive systems under study. In groups 3 and 6, this procedure was performed after applying and polymerizing the adhesive systems.

2.6 Adhesive system application and restorative procedures

For the Adper Scotchbond Multi-Purpose adhesive system: acid application for 15 seconds, washing with water for 30 seconds, drying, primer application, mild air spraying for 5 seconds, adhesive application, and light-curing for 10 seconds. For the Single Bond Universal adhesive system: adhesive layer application for 15 seconds, mild air spraying for 5 seconds, new adhesive layer application, air drying for 5 seconds, and light-curing for 10 seconds.

After polymerizing the adhesive system, a rubber matrix with a central perforation of 3 mm in diameter and 5 mm in height was placed in the adhesive area to facilitate the insertion of the Opallis A3 color nanohybrid restorative composite (FGM LTDA, Joinville, SC, Brazil). The composite was inserted in 2-mm increments aided by a Suprafil spatula and cured for 20 seconds according to the manufacturer's instructions with the Radii-cal curing apparatus (SDI Brasil Indústria e Comércio LTDA, São Paulo, SP, Brazil) at a power of 1200 mw/cm^2. After 24 hours, the specimens were subjected to the mechanical shear strength test.

2.7 Mechanical shear strength test

The sequence of specimen fractures was randomly performed after a draw. The tests were performed in an INSTRON 33R5567 universal testing machine (Instron, Canton, Massachusetts, USA) with a 200-kg load cell adjusted for the speed of 0.5 mm/min. The load was applied with a chisel with a 0.5-mm wide active tip positioned flush with the base of the composite cylinder, as close as possible to the adhesive interface. The load required for fracturing each specimen was expressed in Newton (N), and the results were transformed into Megapascal (MPa) with the formula: P=F/A and submitted to the statistical analysis.

2.8 Statistical analysis

Statistical analyses were performed with the R Project 3.4.2 (R Foundation for Statistical Computing, Vienna, Austria) and IBM SPSS 22 (IBM Corporation, Armonk-NY, United States) software. The Shapiro-Wilks test assessed data normality. Considering the deviation from normality, non-parametric tests were used to assess differences among the groups. The Mann-Whitney test was used for comparing two groups. The Kruskal-Wallis test was performed for analyzing more than two groups and, if there was a significant difference among the groups in the two-by-two analysis (post-hoc), the Dunn test was applied. A 5% significance level was used.

3. Results and Discussion

Table 2 presents the mean values and standard deviations of all groups and the maximum and minimum values of each group. The highest median (30.27 ± 0.54) was recorded for the group that received argon plasma after the application of the Adper Scotchbond Multi-Purpose (SMPPD) adhesive system, and the lowest median (22.66 ± 1.84) was found in the group that received the Single Bond Universal adhesive system without plasma (SBUSP).

Table 3 compares the values obtained between the control groups (which did not receive argon plasma) and

Table 2. Mean, median, standard deviation, minimum/maximum value, and the number of samples for each group tested.

Group	Mean	Median	Standard deviation	Minimum	Maximum
SMPSP	26.74	26.51	0.88	25.27	28.30
SMPPA	29.25	29.22	0.44	28.19	30.07
SMPPD	30.20	30.27	0.54	29.46	31.00
SBUSP	22.46	22.66	1.84	19.71	26.51
SBUAP	28.20	28.33	0.78	26.55	29.22
SBUPD	29.61	29.32	0.91	28.13	31.14
Total	27.74	28.79	2.80	19.71	31.14

the groups that received plasma. According to the Mann-Whitney test ($p<0.05$), the groups that received plasma had higher resistance values (29.30 MPa) than the control groups (25.54 MPa). Table 4 shows the results of the two adhesive systems without plasma application (control group). The Adper Scotchbond Multi-Purpose group showed bond strength values significantly higher than the Single Bond Universal group, whose medians were 26.51 MPa and 22.66 MPa, respectively.

Different letters indicate significant differences between the groups (Mann-Whitney, $p<0.05$). n = 90.

Table 5 shows the results of the influence of plasma application before or after the adhesive system. The results of the Kruskal-Wallis test ($p<0.05$) rejected the null hypothesis that the groups have equal strength values. In both adhesive systems, bond strength values were higher when applying plasma after the adhesive system.

Table 6 shows the comparison of the three treatments tested for the Adper Scotchbond Multi-Purpose adhesive system. The results of the Kruskal-Wallis test ($p<0.05$) rejected the null hypothesis that the groups have equal strength values. The two-by-two comparison with the Dunn test showed higher values for Scotchbond with plasma after > plasma before > no plasma.

Table 7 shows the comparison of the three treatments tested for the Single Bond Universal adhesive system. The results of the Kruskal-Wallis test ($p<0.05$) rejected the null hypothesis that the groups have different strength values. The two-by-two comparison with the Dunn test showed the highest values in the group that received plasma after applying the adhesive system (SBUPD), followed by groups SBUPA and SBUSP.

Forming a dense, homogeneous, and uniform hybrid layer, regardless of thickness, is crucial for strong and lasting adhesion. Thus, the adhesive must completely infiltrate the demineralized dentin, otherwise, porosities or nanometric defects are formed within the hybridized area, facilitating the infiltration of oral fluids and bacterial enzymes that degrade the restoration over time and decrease bond strength and durability[14]. Several factors inherent to the physicochemical structure of the adhesive, the application techniques, and the intrinsic properties of adhesives can affect this diffusion. Considering the tendency to use simpler and faster systems that are less susceptible to operator errors and attempting to eliminate the inconveniences of the total acid etching technique caused by acid washing and substrate drying, self-etching systems were developed with increasing concentrations of acidic monomers in their composition[15]. The low pH values of these systems allow them to partially diffuse through the dentin slurry, reaching and superficially demineralizing the underlying intact dentin until dissolution products buffer its acidity[15]. Acidic monomers decalcify the adhesive and simultaneously open channels that facilitate the diffusion of resin monomers within the dentin, forming a thin hybridized complex. However, there are no discrepancies between the depths of monomer demineralization and infiltration, providing satisfactory initial bond strength values[16,17], as observed in this study for the Single Bond Universal system in the control group.

Considering the more uniform interdiffusion zone, it was expected that the adhesion values of the self-etching system evaluated would be the highest among the control groups. However, the results obtained for the previously acid-etched Scotchbond Multi-Purpose conventional three-step system were statistically higher than those of the Single Bond Universal system. This may have occurred because the SBMP system is less sensitive to substrate moisture variations due to the presence of water in its composition, which can rehydrate the collapsed collagen, recover its original

Table 3. Values, in MPa, of control groups and groups that received plasma.

Group	Median	Standard deviation
With plasma	29.30 A	1.00
Without plasma	25.54 B	2.60
Total	28.79	2.80

Table 4. Shear strength values, in MPa, of the two adhesive systems without plasma application.

Group	Median	Standard deviation
SBMPSP	26.51 A	0.88
SBUSP	22.66 B	1.84
Total	25.54	2.60

Different letters indicate significant differences between the groups (Mann-Whitney, $p<0.05$). n = 30.

Table 5. Shear strength values, in MPa, of the two adhesive systems with plasma application.

Group	Median	Standard deviation
SMPPA	29.22 B	0.44
SMPPD	30.27 A	0.54
SBUPA	28.33 C	0.78
SBUPD	29.32 B	0.91
Total	29.30	1.00

Different letters indicate significant differences between the groups (Mann-Whitney, $p<0.05$). n = 60.

Table 6. Comparison of shear strength values, in MPa, for the three treatments with the Adper Scotchbond Multi-Purpose adhesive system.

Group	Median	Standard deviation
ASMPSP	26.51 C	0.88
ASMPPA	29.22 B	0.44
ASMPPD	30.27 A	0.54
Total	29.22	1.61

Different letters indicate significant differences between the groups (Dunn test, $p<0.05$). n = 45.

Table 7. Comparison of shear strength values, in MPa, for the three treatments with the Single Bond Universal adhesive system.

Group	Median	Standard deviation
SBUSP	22.66 C	1.84
SBUPA	28.33 B	0.78
SBUPD	29.32 A	0.91
Total	28.29	3.36

Different letters indicate significant differences between the groups (Dunn test, $p<0.05$). n = 45.

framework in dry dentin situations after acid washing, and present hydrophilic monomers with a high affinity for the substrate under higher moisture conditions[12]. Although this system does not have a simultaneous hybridization to dentin demineralization, it is less sensitive to errors caused by the application technique. Therefore, reliable adhesion values can be obtained in different substrate moisture conditions, among other characteristics, which qualifies this system as the gold standard for dental adhesion[12].

Additionally, the narrow thickness of the interdiffusion zone formed by the Single Bond Universal system results in a hybrid layer with a high elastic modulus, reducing its flexibility and ability to absorb the stresses generated when polymerizing the restorative composite, forming more cracks that propagate fractures in the joint area during the mechanical test. In turn, the SBMP system uses a more viscous adhesive composed of Bis-GMA and HEMA after the primer, which allows forming a more elastic layer with a higher potential for absorbing the forces generated during polymerization shrinkage[18]. It is also worth noting that the potentially acidic medium promoted by the self-etching primer makes it hard to convert resin monomers into polymers[11], and incorporating the smear layer in the hybrid complex of self-etching systems causes a weak bond adherence[17].

These findings reject the null hypothesis that the composition of adhesive systems does not influence dentin bond values, considering the values of the SBMP system were higher than those of the SBU system in the three variables studied. There have been several attempts to create a defect-free hybrid layer. Several authors have proposed different types of dentin pretreatments to increase the durability of adhesive restorations. Dentin deproteinization, collagen cross-linking agents, and laser irradiation are examples of poorly consolidated attempts to improve adherence[19,20].

Recent efforts have been focused on developing a technique to electrically or chemically modify the dentin surface. Methods have been used to increase the permeability and wettability of this substrate and facilitate the penetration and absorption of adhesive agents[21]. Thus, the non-thermal plasma application technique has attracted considerable interest and has been extensively used to modify the surfaces of biomaterials[22].

This study used non-thermal argon plasma because it is inexpensive compared to other noble gases, and the application temperature is close to the human body temperature, (lower than 40°C at the time of application), which allows using it *in vivo*. Non-thermal argon plasma also presented favorable adhesive strength results in the articles evaluated[6,10]. The time of 30 seconds was chosen due to its clinical application feasibility.

Studies have suggested that non-thermal plasma can increase interfacial bond strength by increasing the surface contact area with the collagen fibers and their hydrophilicity, allowing a higher interaction with the adhesive and penetration into the substrate. This reduces defects and voids at the interface, forms longer resin extensions, and increases the conversion of resin monomers into polymers[23-25], corroborating the findings of this research, in which bond strength values were significantly higher for both adhesive systems when treating the dentin substrate with plasma. Therefore, the null hypothesis that the treatment with argon plasma does not interfere with the adhesive strength of the systems used was rejected.

This behavior was verified for both adhesive systems when the plasma was applied before the adhesives to dentin. As dentin is a substrate rich in organic matter, the contact angle between this substrate and a liquid is higher than that of enamel, impairing the adhesive process[10,26]. The results obtained with non-thermal argon plasma indicate that this gas is can make the dental surface more hydrophilic, considerably reducing the dentin contact angle, and increasing wettability and consequent penetration of the adhesive system[10]. This occurs because plasma breaks down and removes the protein content of hydrocarbons from the dentin surface. This is associated with the stiffening effect on the hybrid layer, inhibiting the enzymatic activity of matrix metalloproteinase (MMP). Another possible explanation is that breaking interfibrillar bonds, such as hydrocarbon bonds, can induce structural changes in the exposed collagen fibers, preventing the collagen networks from collapsing, protecting them, and inhibiting MMP enzymes, thus improving bond durability. Therefore, reducing the amount of organic matter increases the amount of mineral content, mainly calcium and phosphate - the main components of hydroxyapatite. This reduces the surface contact angle, favoring the close contact of the adhesive and its penetration[27-29].

This behavior becomes more significant when using self-etching adhesive systems because of the absence of a pre-etching step, therefore, not removing the smear layer from this surface. The presence of the dentin slurry reduces the surface energy of the substrate, making it less receptive to adhesion. It also works as a physical barrier to monomer penetration, further impaired by the increased thickness of this debris layer, reducing primer acidity and hindering monomer diffusion in the underlying dentin[30]. Applying plasma directly to the dentin without pre-etching in the groups that received the SBU system may have changed the composition of the dentin slurry, increasing the surface energy of the substrate and favoring bonding[30], which was translated by the significant difference in the numerical results obtained for the adhesive.

The strong chemical and physical bonds promoted by plasma increase mechanical strength values, as verified in this study, in which shear strength values were significantly higher for the groups with previous plasma applications. The null hypothesis that applying non-thermal argon plasma before the adhesive system does not influence the shear strength values of the adhesives evaluated was rejected. Despite the increase in bond strength values with the prior dentin treatment with argon plasma, these values were significantly lower than those obtained with the application of the gas after substrate hybridization in both adhesive systems evaluated. This possibly occurred due to using deep dentin as the substrate for adhesion, considering that morphological, structural, and compositional differences occur at different depths of the dentin substrate[22,31,32]. Some studies[33,34] report that deep dentin has a lower mineral content and amount of collagen fibers caused by the increase in density, width, and area occupied by dentinal tubules, which may have impaired plasma performance when applied before the adhesive.

The benefits of this treatment are closely related to the existing amount of tissue, especially collagen fibers[32-34].

However, the higher dentin depth did not influence the values obtained with plasma application after the hybrid layer formation, as they were the highest adhesion values for both systems evaluated. This occurred because *in vitro* adhesion is not affected by increased moisture in the deep substrate, which can cause adhesive system dilution, phase separation, and reduction of the degree of monomeric conversion[12] – the main action mechanism of plasma on the hybridized tooth surface[33]. The null hypothesis that applying non-thermal argon plasma gas after hybridization does not influence the bond strength values of the systems evaluated was rejected. The acidic monomers in these systems react with the initiating amines, reducing their concentration and polymerization reaction, which can negatively affect adhesion[10,11]. However, applying plasma after the adhesive may have minimized this problem due to the additional polymerization, as seen in the values of the Single Bond Universal system in this experimental condition.

Also, self-etching systems consist of a certain amount of water, as in the single-bottle system evaluated in this study, which contains 20%. Water is essential for ionizing acid monomers, but it can be an interference factor, reducing the photopolymerization of adhesives by diluting its components. However, the water contained in acidic adhesives does not have a deleterious effect when using plasma, considering that an appropriate amount of water can facilitate the injection of free radicals into the plasma and increase the propagation of the monomeric chain, thus increasing conversion[30-32].

4. Conclusions

Applying non-thermal argon plasma for 30 seconds improved the bond strength of deep dentin. Non-thermal argon plasma applied to deep dentin before the adhesives significantly increased the bond strength of both adhesive systems, which ensured higher wettability and adhesion.

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