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Effect of Gallic Acid Pretreatment and Application Mode on Dentin Bond Strength of a Universal Adhesive System After Thermal Aging: An In Vitro Study

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Abstract

Background: This in vitro study evaluated the effects of two different adhesive application approaches (total-etch and self-etch) and gallic acid (GA) pretreatment on the dentin microshear bond strength (μSBS) of a universal adhesive system. Bond strength was assessed both before thermal aging and following aging procedures simulating approximately 1 and 5 years of clinical service. **Materials and Methods:** One hundred twenty intact human incisors were allocated to experimental groups according to the adhesive strategy, presence or absence of gallic acid (GA) pretreatment, and thermocycling regimen (0, 10,000, or 50,000 cycles). A universal adhesive system (G-Premio BOND) in combination with a nanohybrid composite resin was applied in accordance with the manufacturers' instructions. Microshear bond strength (μSBS) was determined using a universal testing device. The obtained data were analyzed by three-way ANOVA and subsequently compared using Tukey's post hoc test at a significance level of 0.05. **Results:** In the total-etch approach, pretreatment with gallic acid (GA) resulted in significantly greater μSBS values than those observed in the corresponding untreated specimens under all aging conditions (no thermocycling: 18.53 ± 0.99 vs. 11.33 ± 0.81 MPa; 1-year: 19.86 ± 0.82 vs. 11.60 ± 0.58 MPa; 5-year: 19.04 ± 0.62 vs. 10.28 ± 0.83 MPa; $p = 0.001$). A comparable trend was noted for the self-etch strategy, where GA application significantly enhanced bond strength compared with the non-treated groups (no thermocycling: 21.70 ± 0.98 vs. 14.19 ± 1.17 MPa; 1-year: 22.60 ± 0.50 vs. 14.94 ± 0.85 MPa; 5-year: 22.32 ± 0.59 vs. 12.94 ± 0.84 MPa; $p = 0.001$). Across all thermocycling conditions, the self-etch mode consistently produced higher bond strength values than the total-etch mode. Thermal aging did not significantly influence μSBS in the GA-treated groups. In contrast, in the absence of GA pretreatment, thermocycling led to a reduction in bond strength, particularly after the 5-year aging protocol. **Conclusions:** Gallic acid pretreatment significantly improved dentin bond strength and contributed to the preservation of bond durability after thermal aging. The highest μSBS values were obtained when the self-etch approach was combined with gallic acid (GA) pretreatment, suggesting that GA may serve as a beneficial adjunct for improving the durability and long-term performance of resin–dentin bonds.

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Keywords: gallic acid; microshear bond strength; self-etch; total-etch

1. Introduction

Adhesive technologies have fundamentally reshaped contemporary restorative dentistry by enabling minimally invasive treatment approaches and improving the retention of resin-based materials. Nowadays, one-step universal adhesive systems are attracting growing attention in clinical practice because they can be used in both total-etch and self-etch approaches. Despite significant technological improvements, maintaining the durability of the resin–dentin interface over time remains a persistent clinical concern [1]. Achieving an ideal bond between composite resin restorative materials and the dentin substrate continues to be challenging, and there is still uncertainty regarding whether the adhesive system can properly infiltrate the collagen framework to create a compatible structure [2]. It has been proposed that hydrolytic degradation, together with the action of dentin matrix enzymes, compromises the tooth–adhesive bond and causes the deterioration of unprotected collagen fibrils [3]. While adequate resin–dentin bonding can often be obtained initially, bond strength typically decreases over time [4,5].

Laboratory-based studies indicate that dentin pretreatment with gallic acid may positively influence bond performance of universal adhesives applied using different etching strategies [6]. The durability of the adhesive interface has been closely associated with the activity of endogenous matrix metalloproteinases, which contribute to collagen disintegration within the hybrid layer [7]. By modulating this enzymatic activity, naturally derived polyphenols have been reported to enhance collagen stability and limit structural degradation, thereby supporting the potential use of GA as a biologically active adjunct in adhesive procedures.

In total-etch applications, the acid used completely removes the smear layer, causing demineralization of mineralized dentin tissues, specifically peritubular and intertubular dentin [8]. On enamel surfaces, acid etching produces macro- and micro-porosities, and the penetration of the hydrophobic bonding agent into these structures creates micromechanical interlocking, which results in the strongest possible attachment to the dental substrate [9]. However, bonding to dentin is inherently more complex and technique-sensitive due to its structural nature [9].

The bonding mechanism of self-etch adhesives involves two components: micromechanical retention and chemical interaction with dental tissues [10]. Micromechanical adhesion results from the partial dissolution of minerals and their replacement by resin monomers [10]. While micromechanical retention helps prevent early bond failure, chemical interactions contribute to stabilizing the interface against degradation over time [10].

Gallic acid (GA), commonly found in grapes and green tea, is known for its strong anti-inflammatory effects [11], antiviral properties [12], and antibacterial activity against microorganisms responsible for dental caries [13]. Moreover, GA has been reported to mimic natural cross-linking processes within dentin, thereby reducing the biological degradation of collagen fibers and enhancing their structural stability, which contributes to improved clinical outcomes in restorative procedures [14]. GA derivatives (catechin gallates and gallocatechins) have been shown to inhibit tissue-degrading enzymes such as MMPs and cathepsins, suggesting that they may offer therapeutic benefits in conditions characterized by excessive matrix degradation [15]. Compared with other collagen cross-linking agents previously investigated in dentin bonding, gallic acid has attracted attention due to its low cytotoxicity, natural origin, and dual ability to enhance collagen stability while inhibiting matrix-degrading enzymes [15]. In the present study, a 1% aqueous GA solution was prepared by dissolving 1 g of gallic acid powder (Sigma Aldrich, St.

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Louis, MO, USA) in 100 mL of distilled water. This solution was applied 10 s prior to adhesive placement. Including this brief, clinically feasible application allows the potential collagen-stabilizing and MMP-inhibitory effects of GA to be realized while considering the acidic/ultra-mild characteristics of universal adhesives, which may influence the interaction between GA and dentin collagen. The adhesive used in this study, G-Premio BOND (GC Corporation, Tokyo, Japan), has a pH of 1.5, and this acidic environment may affect the interaction between gallic acid (GA) and dentin collagen as GA's potential collagen-stabilizing and MMP-inhibitory effects could be modulated by the adhesive's pH. These characteristics position gallic acid as a potential option for enhancing the longevity of the resin–dentin interface while maintaining its clinical usability. To approximate intraoral thermal challenges under controlled conditions, specimens are often subjected to cyclic temperature changes in a laboratory setting. Such repeated thermal fluctuations can promote interfacial fatigue and facilitate degradation of the resin–dentin bond [16].

Understanding whether simple modifications such as etching mode selection and gallic acid pretreatment can improve the long-term stability of dentin bonding may provide clinicians with practical approaches for enhancing the longevity of adhesive restorations.

Earlier investigations have reported improvements in immediate bond strength following gallic acid application, as well as its ability to interfere with collagenolytic enzyme activity. However, information remains scarce concerning its durability when combined with different adhesive protocols and subjected to prolonged thermal aging. In particular, it remains unclear whether the potential collagen-stabilizing and MMP-inhibitory effects of GA can effectively preserve dentin bond strength when combined with different etching modes and subjected to long-term thermocycling that simulates clinical aging. Therefore, further investigation is required to clarify whether GA pretreatment provides additional durability benefits depending on the adhesive application protocol and aging conditions.

The present investigation evaluated dentin microshear bond strength of a universal adhesive used in total-etch or self-etch mode, with and without gallic acid pretreatment, both before aging and after thermocycling protocols corresponding to 10,000 and 50,000 cycles. The first null hypothesis was that neither the adhesive application mode (total-etch or self-etch) nor the use of gallic acid before adhesive application would influence dentin microshear bond strength. The second null hypothesis was that different thermal aging conditions, including no thermocycling, 1-year thermocycling (10,000 cycles), and 5-year thermocycling (50,000 cycles), would not affect dentin microshear bond strength.

2. Materials and Methods

Ethical clearance for this investigation was obtained from the Clinical Research Ethics Committee of Gazi University Faculty of Dentistry (Protocol No.: E-77082166-604.01-1395056). All laboratory procedures were performed at the Department of Restorative Dentistry, Gazi University. Sample size determination was carried out using G*Power software (https://g-power.apponic.com/) through an a priori power analysis. Based on an anticipated effect size of 0.50, an alpha value of 0.05, and a power of 80%, the minimum required sample size was calculated as 10 specimens per group.

2.1. Preparation of Specimens

A total of 120 intact human incisors, extracted for periodontal reasons, were included in this study. The roots were sectioned 2 mm below the cemento-enamel junction, and the buccal enamel was removed using a precision cutting device (Brillant 210 Precision Cutting Machine, Ata Quality Assured, Liverpool, UK) to expose the dentin. Each tooth was then embedded in cold-cure acrylic resin inside polyvinyl chloride (PVC) cylinders, with

the buccal surface facing upward. The dentin surfaces were standardized by polishing for 30 s with 600-grit silicon carbide paper to produce a uniform smear layer.

The samples were randomly assigned to twelve groups (n = 10) (Figure 1, Table 1). The properties of the adhesive and composite materials used are summarized in Table 2. A 1% gallic acid (GA) solution was prepared by dissolving 1 g of GA powder (Gallic acid, Sigma Aldrich, USA) in 100 mL of distilled water.

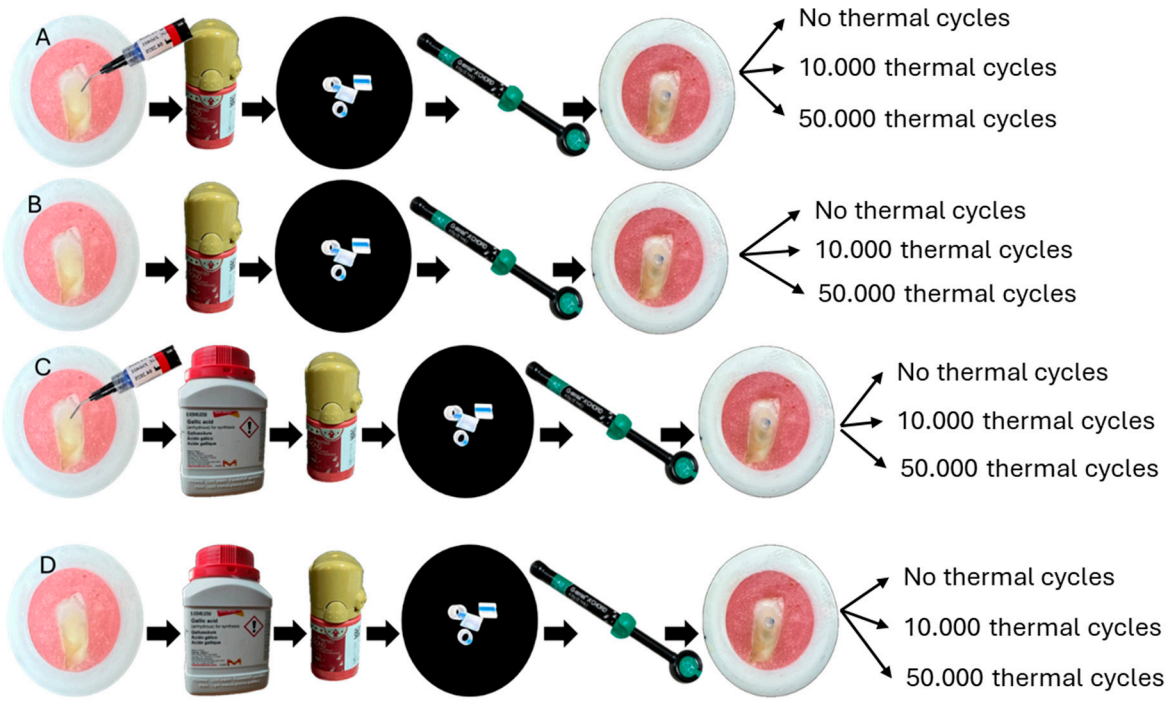


Figure 1. (**A**) Total-etch mode application, (**B**) Self-etch mode application, (**C**) Total-etch mode application in combination with gallic acid, (**D**) Self-etch mode application in combination with gallic acid.

Table 1. Experimental groups according to adhesive application mode, gallic acid pretreatment, and thermocycling conditions.

Group (n = 10)	Adhesive Application Mode	Gallic Acid Pretreatment	Thermocycling
1	Total-etch	No	None
2	Self-etch	No	None
3	Total-etch	Yes	None
4	Self-etch	Yes	None
5	Total-etch	No	1-year (10,000 cycles)
6	Self-etch	No	1-year (10,000 cycles)
7	Total-etch	Yes	1-year (10,000 cycles)
8	Self-etch	Yes	1-year (10,000 cycles)
9	Total-etch	No	5-year (50,000 cycles)
10	Self-etch	No	5-year (50,000 cycles)
11	Total-etch	Yes	5-year (50,000 cycles)
12	Self-etch	Yes	5-year (50,000 cycles)

Table 2. Materials, manufacturers, components, and type of material (MDP, 10-Methacryloyloxydecyl dihydrogen phosphate; 4-MET, 4-Methacryloyloxyethyl trimellitate acid; MEPS, Methacryloyloxyethyl phenyl hydrogen succinate; Bis-MEPP, Bisphenol-A diglycidyl ether methacrylate phosphate; UDMA, Urethane Dimethacrylate).

Material	Manufacturer/Lot Number	Main Components	Type of Material
G-Premio Bond	GC, Tokyo, Japan/ 2411264	MDP, 4-MET, MEPS, methacrylate monomer, acetone, water, initiator, silica (pH = 1.5)	Universal adhesive
G-ænial A'CHORD	GC, Tokyo, Japan/ 220523A	CERASMART, Bis-MEPP, UDMA/3 μm	Nanohybrid composite

In Groups 1, 5, and 9, the universal adhesive system was applied in the total-etch mode. The dentin surfaces were first treated with 37% phosphoric acid gel (K-etchant Gel, Kuraray Noritake Dental, Tokyo, Japan) for 15 s, and then rinsed for an additional 15 s and gently air-dried. The adhesive (G-Premio BOND, GC Corporation, Tokyo, Japan) was applied with a microbrush for 10 s, lightly air-thinned, and cured for 10 s using an LED light-curing unit (D-Light Pro, GC, Tokyo, Japan) with a wavelength of 430–480 nm and an intensity of 1200 mW/cm^2, following the manufacturer's guidelines.

For Groups 2, 6, and 10, in which the adhesive was applied in self-etch mode, the adhesive (G-Premio BOND) was directly applied with a microbrush for 10 s, lightly air-thinned, and light-cured for 10 s. The same light-curing protocol was applied to all other groups.

In Groups 3, 7, and 11, where gallic acid pretreatment was combined with total-etch application, the dentin surfaces were initially etched with 37% phosphoric acid gel for 15 s and rinsed for 15 s. A 1% GA solution was then applied for 10 s using a microbrush, followed by rinsing with water for 10 s and gentle air-drying. Finally, the adhesive (G-Premio BOND) was applied for 10 s, lightly air-thinned, and cured for 10 s.

In Groups 4, 8, and 12, which involved gallic acid pretreatment followed by the self-etch adhesive protocol, a 1% GA solution was applied to the dentin surface with a microbrush for 10 s, and then rinsed for 10 s and gently air-dried. The adhesive (G-Premio BOND) was subsequently applied for 10 s, lightly air-thinned, and light-cured for 10 s.

A transparent tube with an internal diameter and height of 2 mm (Tygon, Norton Performance Plastic Co., Akron, OH, USA) was positioned on the dentin surface, and the mold was filled with a nanohybrid composite resin (G-ænial A'CHORD, GC Corporation, Tokyo, Japan).

2.2. Thermocycling Procedure

The number of thermocycles applied in this study was determined based on previous research, which considers 10,000 cycles roughly equivalent to one year of clinical service and 50,000 cycles as representing extended aging conditions [17]. Prior to aging, all prepared specimens were stored in a humidified incubator at 37 °C for 24 h. Groups 1–4 were not subjected to thermocycling, whereas Groups 5–8 underwent 10,000 cycles and Groups 9–12 underwent 50,000 cycles, simulating approximately one and five years of intraoral aging, respectively. The specimens were carefully placed in the basket compartment of a thermocycler (Thermocycler, SD Mechatronik, Feldkirchen-Westerham, Germany) without contacting one another. The thermocycler was calibrated according to the manufacturer's guidelines before use. Aging was carried out by alternately immersing the specimens in water baths at 5 °C and 55 °C, with a dwell time of 20 s in each bath and a 10-s transfer interval between baths. These parameters were selected based on previous studies [18] to simulate clinical aging and ensure reproducibility.

2.3. Micro-Shear Bond Strength Test Application

The micro-shear bond strength of the specimens to dentin was evaluated using the micro-shear testing method. After being stored in distilled water at 37 °C for 24 h in an incubator, the specimens underwent thermocycling and were then tested with a universal testing machine (Shimadzu IG-IS, Kyoto, Japan) at a crosshead speed of 1 mm/min. The forces recorded in Newtons were converted to megapascals (MPa) by dividing by the bonded surface area. The bonded area was calculated based on the internal diameter of the tube (2 mm) using the following formula:

$$\text{Bond strength (MPa)} = \frac{\text{Force (N)}}{\pi \times (d/2)^2} \quad (1)$$

where d represents the internal diameter of the tube. Before testing, the universal testing machine was calibrated following the manufacturer's instructions to guarantee precise and consistent measurements. The obtained data were documented for statistical analysis. Although μSBS standards recommend a bonded area diameter of less than 1 mm, a 2 mm diameter was used in the present study due to practical considerations related to specimen preparation and handling. Previous studies have shown that diameters larger than 1 mm can still be acceptable for micro-shear testing without compromising the validity of the results [19].

2.4. Statistical Analysis

Statistical analyses were performed using IBM SPSS Statistics version 27. Data normality was evaluated using the Kolmogorov–Smirnov and Shapiro–Wilk tests, which confirmed that all variables followed a normal distribution. To assess the effects of application method, gallic acid pretreatment, and thermocycling on bond strength, a three-way ANOVA was conducted, followed by Tukey's post hoc test. A *p*-value of less than 0.05 was considered statistically significant.

3. Results

When comparing the total-etch and self-etch application modes, the gallic acid–treated groups exhibited significantly higher mean microshear bond strength (μSBS) values than the untreated groups, irrespective of the thermocycling conditions (p = 0.001) (Table 3).

Table 3. Evaluation of the combined effects of adhesive application mode, gallic acid, and thermocycling on bond strength.

		With Gallic Acid	Without Gallic Acid	
Adhesive Application Mode	**Thermocycling**	**Mean ± SD**	**Mean ± SD**	***p***
Total-etch	No thermocycling	18.53 ± 0.99 ^{A}	11.33 ± 0.81 ^{A}	0.001 *
	1-year thermocycling	19.86 ± 0.82 ^{B}	11.60 ± 0.58 ^{A}	0.001 *
	5-year thermocycling	19.04 ± 0.62 ^{AB}	10.28 ± 0.83 ^{B}	0.001 *
	p	0.004 *	0.001 *	
Self-etch	No thermocycling	21.70 ± 0.98 ^{A}	14.19 ± 1.17 ^{A}	0.001 *
	1-year thermocycling	22.60 ± 0.50 ^{B}	14.94 ± 0.85 ^{A}	0.001 *
	5-year thermocycling	22.32 ± 0.59 ^{AB}	12.94 ± 0.84 ^{B}	0.001 *
	p	0.028 *	0.001 *	

Three-Way ANOVA Test * $p < 0.05$. Different uppercase letters denote statistically significant differences among thermocycling groups within the same adhesive application mode.

Within the gallic acid-treated groups, thermocycling had a significant impact (p = 0.004), with the 1-year thermocycling group showing higher bond strength than the non-

thermocycled specimens (p = 0.003). No significant differences were found among the other thermocycling conditions (p > 0.05).

In the groups without gallic acid, thermocycling also affected bond strength (p = 0.001). Specifically, 5-year thermocycling led to a significant reduction in μSBS compared to both the non-thermocycled group (p = 0.011) and the 1-year thermocycled group (p = 0.001). No significant differences were observed among the other comparisons (p > 0.05).

Across all thermocycling conditions, the self-etch application method consistently showed significantly higher mean bond strength than the total-etch mode in both gallic acid-treated and untreated groups (p = 0.001) (Figure 2).

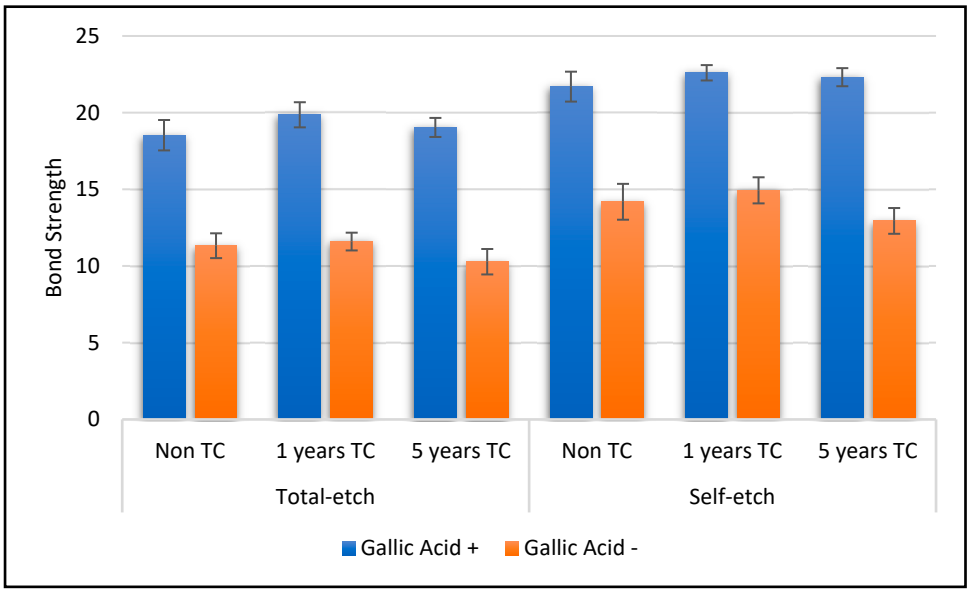


Figure 2. Distribution of bond strength values according to adhesive application mode, gallic acid application, and thermocycling conditions (TC; Thermocycling).

4. Discussion

The findings of this study indicated that the self-etch application mode resulted in significantly higher microshear bond strength values compared to the total-etch method. Additionally, pretreatment with gallic acid significantly enhanced bond strength, leading to the rejection of the first null hypothesis. Extended thermocycling negatively affected bond strength, resulting in the rejection of the second null hypothesis.

In total-etch systems, the smear layer is removed using 30–40% orthophosphoric acid, exposing the collagen network. The subsequently applied resin penetrates the dentinal tubules and intercalates between the collagen fibers, forming the hybrid layer after polymerization [20]. In contrast, self-etch adhesives contain acidic monomers that allow them to simultaneously demineralize and penetrate dentin without the need for a separate rinsing step. These adhesives also interact with the smear layer, incorporating it into the hybrid layer [21]. To enhance clinical versatility, “universal” or “multimode” adhesives have been developed, which can be applied using either the self-etch or total-etch approach.

Takamizawa et al. [22] investigated the dentin bond strength of universal adhesives applied with different techniques and reported a reduction in bond strength for Clearfil SE Bond when used in the etch-and-rinse mode. Similarly, Valizadeh et al. [23] assessed the dentin microshear bond strength of Single Bond, Scotchbond Universal, and Clearfil SE Bond using both etch-and-rinse and self-etch methods, finding that the highest microshear bond strength values were observed with the self-etch approach, although the differences were not statistically significant. Due to the lower degree of dentin

demineralization, the authors recommended the self-etch technique [23]. Consistent with these findings, the present study showed that, under all conditions—including no thermocycling as well as 1-year and 5-year thermocycling—and in both gallic acid-treated and untreated groups, the mean bond strength of the self-etch groups was significantly higher than that of the total-etch groups ($p = 0.001$; $p < 0.05$).

The hybrid layer is composed of type I collagen fibrils and proteoglycans that are encapsulated within polymer chains [24,25]. These collagen fibrils are particularly vulnerable to degradation in the presence of microleakage, which is mainly caused by mechanical and hydraulic fatigue within the restoration. Furthermore, the activation of matrix metalloproteinases (MMPs) and cysteine cathepsins contributes to the accelerated degradation of collagen fibrils [26].

Matrix metalloproteinases (MMPs) present in dentin can also be activated by the acidic properties of adhesive systems. Nishitani et al. [27] and Tay et al. [28] reported that mild acids are capable of activating dentinal MMPs. Besides MMPs, acidic monomers can also trigger cysteine cathepsins, resulting in MMP–cathepsin interactions that are believed to contribute to the breakdown of collagen within the hybrid layer [29]. This enzymatic activity can result in gradual bond deterioration over time. In total-etch systems, insufficient resin penetration into the collagen matrix is often observed, leaving collagen fibrils exposed and potentially hastening the degradation process [30]. Mazzoni et al. [31] demonstrated that etch-and-rinse systems, and Nishitani et al. [27] showed that self-etch adhesives, can reactivate gelatinases (MMP-2 and MMP-9) and collagenases in demineralized dentin. However, because self-etch systems induce a shallower degree of demineralization, this effect is considered less pronounced. In addition to its ability to cross-link collagen, gallic acid has been shown to inhibit matrix metalloproteinases, particularly MMP-2 and MMP-9 [32,33]. Previous research has indicated that GA decreases the expression and activity of these gelatinases by modulating intracellular signaling pathways, such as the MAPK and NF-κB pathways [32,33]. Gallic acid (GA) may enhance bond durability by suppressing MMP-2 and MMP-9 activity, which reduces enzymatic breakdown of exposed collagen fibrils in the hybrid layer. From a broader mechanistic perspective, the enhanced performance of the self-etch approach and the positive effect of GA seem to be closely linked, especially under thermocycling conditions. The self-etch strategy produces a more limited depth of demineralization, reducing the amount of exposed collagen susceptible to enzymatic degradation. In contrast, total-etch systems create a deeper demineralized zone, which increases the risk of incomplete resin infiltration and subsequent collagen breakdown. Within this framework, GA may exert a dual protective effect by enhancing collagen cross-linking and suppressing MMP-2 and MMP-9 activity. Under thermocycling-induced hydrolytic stress, these protective mechanisms likely contributed to preservation of the hybrid layer integrity, explaining why GA-treated specimens demonstrated improved long-term bond stability compared with untreated groups. In the present study, the GA solution was rinsed after 10 s of application. This rinsing was intentionally performed to focus on surface modification of dentin collagen rather than incorporation into the hybrid layer. This approach was chosen to simulate a clinically feasible protocol and to evaluate whether short-term surface interaction with GA is sufficient to enhance bond durability. Bedir et al. [34] reported that GA binds to collagen cleavage sites and reduces collagen degradation. It is noteworthy that gallic acid was rinsed after 10 s of application, yet a significant improvement in bond durability was observed. This finding suggests that even short-term exposure may allow GA to interact with exposed collagen fibrils through hydrogen bonding and potential cross-linking mechanisms. These interactions may increase the resistance of collagen to enzymatic breakdown without the need for long-term retention of GA within the hybrid layer. Consequently, the effectiveness of GA in the present study seems to be related to its capacity to induce rapid

biochemical changes at the surface level rather than to its prolonged chemical incorporation into the adhesive interface. This property may have contributed to the higher microshear bond strength values seen in the GA-treated groups compared with the controls; however, it is primarily associated with the long-term stability of the bond. Thus, the positive effect of gallic acid seems to be primarily linked to the preservation of long-term bond durability rather than simply increasing immediate bond strength. This finding underscores the protective role of gallic acid against prolonged hydrolytic and enzymatic degradation at the resin–dentin interface.

Ghanbarian et al. [6] examined the effect of different concentrations of GA applied to the dentin surface on the shear bond strength of composite resin and reported that a 1% GA solution resulted in the greatest improvement in bond strength. Based on these findings, a 1% GA solution was used in the present study. The results demonstrated that, for both total-etch and self-etch application modes, the GA-treated groups exhibited significantly higher mean bond strength than the untreated groups under non-thermocycled, 1-year, and 5-year thermocycling conditions ($p = 0.001$).

Artificial aging methods allow for the evaluation of potential long-term degradation of restorative materials in a short period by simulating the oral environment [35]. Several artificial aging methods have been reported in the literature, such as storage in artificial saliva, immersion in acidic solutions at 37 °C, water storage, and thermocycling [35,36]. Although some authors have favored water aging protocols [37,38], many studies—including the present one—utilized thermocycling [37,38]. Özcan et al. [39] indicated that thermocycling is more effective than other aging techniques in simulating the aging process of composite resins and produces more demanding conditions for composite restorations.

In thermocycling procedures, parameters such as temperature range, dwell time, and number of cycles can influence bond strength results; among these variables, the number of cycles is considered the most critical factor [40]. Gale et al. [41] proposed that, given that restorations are subjected to approximately 20–25 temperature changes per day intraorally, 10,000 thermocycles may represent roughly one year of clinical service for composite restorations. Based on this assumption, 10,000 and 50,000 cycles were included as variables in the present study. Özcan et al. [39] and Rinastiti et al. [42] reported that even 5000 thermal cycles can affect the bond strength of composite materials. Consistent with these findings, the present study showed that, in the absence of gallic acid pretreatment, specimens subjected to 5-year thermocycling exhibited significantly lower bond strength compared with both the non-aged group ($p = 0.011$) and the 1-year thermocycled group ($p = 0.001$) ($p < 0.05$). These results indicate that gallic acid application has a positive effect on the long-term bond strength of restorative materials.

The findings of this in vitro study align with results reported in several clinical and in vivo investigations. Self-etch adhesives have been shown to provide favorable long-term dentin bond durability in clinical trials, consistent with our observation of higher microshear bond strength values compared with those of total-etch systems [43,44]. Moreover, preliminary clinical studies suggest that collagen cross-linking agents, including gallic acid derivatives, can enhance the longevity of adhesive restorations by stabilizing the hybrid layer and limiting enzymatic degradation [34]. Considering that 10,000 and 50,000 thermocycles approximate 1 and 5 years of clinical service, respectively [41], the improved bond strength observed in GA-treated groups may have meaningful clinical relevance. Nonetheless, caution is warranted when extrapolating these in vitro results to the oral environment, as factors such as occlusal load, saliva composition, enzymatic activity, and microbial biofilm were not simulated in this study.

Although the results were significant, several limitations of this study should be considered. First, as an in vitro study, the experimental design—despite incorporating

thermocycling to mimic aging—cannot entirely reproduce the complex biological, mechanical, and chemical conditions present in the oral environment. Variables such as occlusal forces, saliva composition, enzymatic activity, and bacterial biofilm formation were not replicated. Second, only one universal adhesive system and a single concentration (1%) of gallic acid were evaluated; therefore, the results cannot be generalized to other adhesive systems or concentrations. Additionally, the evaluation was limited to microshear bond strength testing, and no morphological or ultrastructural analysis (e.g., SEM evaluation of the hybrid layer) was performed to further clarify the underlying mechanisms. Finally, long-term clinical performance cannot be directly extrapolated from laboratory aging protocols.

5. Conclusions

Within the scope of this study, the primary finding is that self-etch universal adhesives used in combination with gallic acid pretreatment significantly improve and maintain dentin microshear bond strength, even after extended thermocycling. Gallic acid treatment effectively mitigated the reduction in bond strength observed in untreated groups after extended aging, highlighting its protective role at the dentin–resin interface. These results indicate that this combined approach is a promising strategy for improving long-term adhesive durability. Future research should focus on clinical trials and long-term in vivo studies to validate these findings, as well as investigate the effects of different concentrations of gallic acid, alternative adhesive systems, and other aging protocols to optimize the clinical applicability of this approach.

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Informed Consent Statement: Written informed consent was obtained from all participants.

Data Availability Statement: The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding author.

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Abbreviations

GA	Gallic Acid
MDP	10-Methacryloyloxydecyl dihydrogen phosphate
4-MET	4-Methacryloyloxyethyl trimellitate acid
MEPS	Methacryloyloxyethyl phenyl hydrogen succinate
Bis-MEPP	Bisphenol-A diglycidyl ether methacrylate phosphate
UDMA	Urethane Dimethacrylate

TC Thermocycling

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