Keywords
Dentin, micro-shear bond strength, RMGIC, FujiCEM evolve, polyacrylic acid, gallic acid.

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Effect of Gallic acid Pretreatment of Dentin Surface on Bonding Performance of Resin-Modified Glass Ionomer Luting Cement: An in Vitro Study

Abstract

Introduction and Aim: For indirect restorations, resin-modified glass ionomer luting cements (RMGICs) are frequently utilised as luting agents. For a hybrid layer, however, bond durability is an issue of concern is the hydrolytic degradation. The study evaluated the effect of gallic acid (GA) pretreatment of dentin surface on the immediate and delayed micro-shear bond strength (μSBS) of Fuji CEM Evolve RMGIC to dentin.

Materials and Method: Sixty sound extracted human maxillary premolar teeth were used in this study, decoronated perpendicular to the long axis of each tooth exposing a flat dentin surface. Teeth were then divided into three main groups (n=20) according to the type of dentin surface pretreatment used: Group I pretreated with polyacrylic acid (PAA) (control group), Group II pretreated with Gallic acid (GA), and Group III pretreated with PAA followed by GA.

Each group was further subdivided into two subgroups (n=10) according to the aging period prior to testing: Subgroup A (Immediate) tested after 24 hours, and Subroup B (Delayed) tested after 3 months. FujiCEM evolve RMGIC was then applied on the treated dentin surface using a specially designed Teflon Mold and shear bond strength was then tested using a universal testing machine. Field-emission scanning electron microscopy (FE-SEM) was used to examine the resin-dentin interface. The type of bond failure was figured out by examining the debonded samples with scanning electron microscopy (SEM). One-way ANOVA test and Tukey's test were used to analyse the collected data.

Results: Group III recorded the highest μSBS at both time points (14.19 ± 2.07 MPa at 24 hours; 13.97 ± 2.07 MPa at 3 months) significantly outperforming Group I and Group II ($p<0.01$). The bond strength of Group I and Group II dropped significantly over time ($p<0.01$), while Group III maintained its high bond strength over time as revealed by the statistically non-significant difference in μSBS between the immediate and delayed subgroups ($p>0.05$). Failure mode analysis showed that GA specimens recorded 100% adhesive failures at both time points, denoting that the bond was not very strong. On the other hand, PAA and PAA+GA specimens showed mixed failure patterns, but the PAA group had a significant shift toward adhesive failure over time.

Conclusion: Dentin surface pretreatment with Gallic acid after conditioning with polyacrylic acid significantly enhanced the shear bond strength of RMGIC luting and provided more durable bonding performance compared to conventional treatment.

1. INTRODUCTION

In the long-term clinical success of indirect restorations, it is necessary to maintain adhesion for mineralized oral tissues to the restorative biomaterials. For this reason, the luting agents should possess sufficient mechanical properties, tolerate intra oral conditions and possess good and long-lasting bond with the tooth structure and the restoration(1).

RMGICs merge the fundamental advantages of both resin composites and conventional GICs, making them excellent choice for restorative applications. Compared with traditional GIC, these materials offer enhanced mechanical properties, sustained fluoride release , and chemical adhesion to tooth structures(2, 3). RMGICs undergo dual-curing process (acid-base reaction and light-activated polymerization) for better clinical outcomes and management (4). In spite of these benefits, they are susceptible to hydrolytic degradation, which could jeopardize their bond strength and mechanical stability. In addition, the resin phase polymerization may induce volumetric contraction, which may lead to marginal gaps, discoloration at the restoration margin, and a reduction in the longevity of the adhesive (3, 5).

Conditioners are frequently implemented prior to the application of GIC or RMGIC in order to enhance adhesion (6). A chemical, known as Polyacrylic acid (PAA), has been proved to be a conditioning agent which is able to demineralize dentin and partly remove the film layer. This process promotes both chemical ionic exchange and micromechanical interlocking. This, however, may reveal the collagen framework and consequently make it more vulnerable to attack by endogenous enzymes which may be present, such as MMPs (7, 8).

Over time, the bond strength would decrease, causing a progressive degradation of bond strength and the durability of the fixed restoration will be affected (9, 10). In recent years, there has been a major paradigm shift towards the bioactive compounds that can modulate the adhesion interface, but at the same time inhibit the degradation of the interface. Natural polyphenols, including gallic acid (GA), 3,4,5-trihydroxybenzoic acid, have been shown to also inhibit MMPs and induce collagen crosslinking, besides having strong antioxidant activity (11). Two primary mechanisms underline GA's stabilizing effect on dentinal collagen. Initially, GA functions as a potent MMP inhibitor, particularly against MMP-2 and MMP-9, by chelating zinc ions at active sites and inducing conformational changes in the enzyme structure (12). Second, by forming intermolecular and intramolecular hydrogen bonds and covalent interactions with proline-rich regions in the collagen molecule (13). GA causes cross-links between the collagen fibrils (intermolecular cross-links) and inside the fibrils (intramolecular cross-links). This dual action has a beneficial effect on the mechanical properties of the demineralized dentin matrix and makes it more resistant over time to enzymatic and hydrolytic degradation. GA may have the potential to stabilize the dentin matrix and thus increase the longevity of the bonding (14, 15). Hence, it seems interesting to test the integration of GA in the adhesive protocol of RMGIC as an alternative to polyacrylic acid.

Epigallocatechin-3-gallate (EGCG) and proanthocyanidins (PAC) are two other natural polyphenols, which have been previously studied as dentin biomodifiers for enhancing the durability of resin-dentin bond (16, 17) . However, GA is a simpler and more stable phenolic acid, and its effect on bonding performance of RMGICs to dentin has not been previously investigated.

As far the authors know, the bonding properties of PAA and GA have never been studied. Hence, the purpose of this study was to compare the effect of dentin surface pretreatment with GA alone and GA with PAA on the micro-shear bond strength (μSBS) of the RMGIC (Fuji CEM Evolve) on dentin after 24 hours and 3 months of aging. Accordingly, the first null hypothesis was tested to see if there is no significant difference between the μSBS of RMGIC and the various surface treatment groups (PA, GA, and their combination). The second null hypothesis was that there is no difference between the immediate and delayed μSBS of RMGIC. The third option was that the third null hypothesis would be true: The failure mode would not be significantly affected by the type of surface treatment.

2. MATERIALS AND METHOD

The main materials utilized in this study beside their chemical composition are listed in Table1.

Fine powder of gallic acid (CAS number 149-91-7) was obtained. A 1% (W/V) Gallic acid solution was made by dissolving 1g of the powder in 100ml of deionised water. After magnetic stirring for 10 minutes, the mixture was briefly sonified, to produce a clear fully-saturated solution (18).

Table (1): The chemical composition of the main materials used in the study

Material	Manufacturer	Composition	
Fuji CEM Evolve GIC	GC Corporation, Japan	Paste cartridge	Paste cartridge
		Fluoro-alumino-silicate glass Hydroxyethyl methacrylate (HEMA) Dimethacrylate&Bis-MEPP Photo-Initiator Inhibitor Silicon dioxide Pigments	PAA Polybasic carboxylic acid Water Initiator Ytterbium fluoride Silicon dioxide
Dentin Conditioner	GC Corporation, Japan	15% polyacrylic acid Liquid monomer C H 2 = C H C O 2 H	
Gallic acid	Sigma-Aldrich Chemie, Steinheim, Germany	1% Gallic acid Polyphenolic organic compound solution 3,4,5 Trihydroxybenzoic acid (C7H6O5)	

2.1 Sample selection

Six hundred teeth of the first premolar of the maxilla were extracted for orthodontic treatment from the private clinics in the city of Baqub'a and Diyala governorate. The study collection of teeth (2025RJK966) followed all the regulations issued by the "Research Ethics Committee of the College of Dentistry at the University of Diyala". The age of the individuals from whom teeth were extracted ranged from 14-30 years.

2.2 Sample preparation

All the teeth were placed in a specially constructed cylindrical silicon mould (19).2 mm apically to the cemento-enamel junction (CEJ) in acrylic blocks. It is manufactured as recommended by the manufacturer, the silicon mold was filled with the acrylic resin cold cure (VERACIL, Colombia). After that, each tooth was decoronated by sectioning the buccal and palatal cusps (1.5 mm) with an electrical diamond saw (Gamberini, Italy) and utilising a lot of water as a coolant. A grinder/polisher machine (MO Pao 160E, China) was then used to complete the cut surface. Standardisation of the smear layer and to obtain a flat and clean surface was the aim of this procedure (20).

2.3 Sample grouping

Three primary groups were created from the tooth specimens: Group I – twenty teeth served as control group pretreated with polyacrylic acid (PAA), Group II – twenty teeth were pretreated with Gallic acid (GA) and Group III – twenty teeth were pretreated with PAA followed by GA. These 2 groups were then split into 2 subgroups, 10 teeth in each group with Subgroups A (Immediate) tested after 24 hours, and Subgroups B (Delayed) tested after 3 months.

2.4 Dentin surface pretreatment

Group I: In Group I the dentin surface was conditioned with 15% PAA (Dentin Conditioner, GC Corporation, Japan) for 10 seconds, applied with a micro brush (Ultradent, USA) and then washed with air water spray and gently air dried to give a 'moist glistering surface' (21).

Group II: In this group, dentin surface was pretreated with 1% Gallic acid, prepared as mentioned formerly, left for 1 minute, then the excess material was removed by a micro brush without rinsing.

Group III: In this group, dentin surface was pretreated using 15% PAA as for Group I, then followed by the application of 1% GA for 1 minute as antioxidant agent and left without rinsing, only removing of the excess material by micro brush (22). Finally, the excess material was blown away to create a glistening and damp surface followed by FujiCEM GIC.

2.5 Cement application

A specially designed split mold to hold the acrylic-embedded tooth specimens and to vertically place standardized polyethylene microtubes (3 mm height × 1 mm internal diameter) was constructed. The mold was made up of two half-circle shapes, 3 mm thick, which were stuck together around the acrylic block and held in place with four fasteners. A removable metal bar with a central bolt attached to the base of the mold to stabilize the assembly, and a removable cover, featuring a 2 mm diameter opening, was placed to guide the specimen surface and accommodate the microtubes. The microtubes were gained by sectioning a 6-FG Nelaton catheter (Fig 1A) (23).

FujiCEM Evolve (GC Corporation, Japan) was dispensed using an automix syringe equipped with an endodontic tip, which was precisely positioned within the lumen of each polyethylene microtube (Fig 1B).

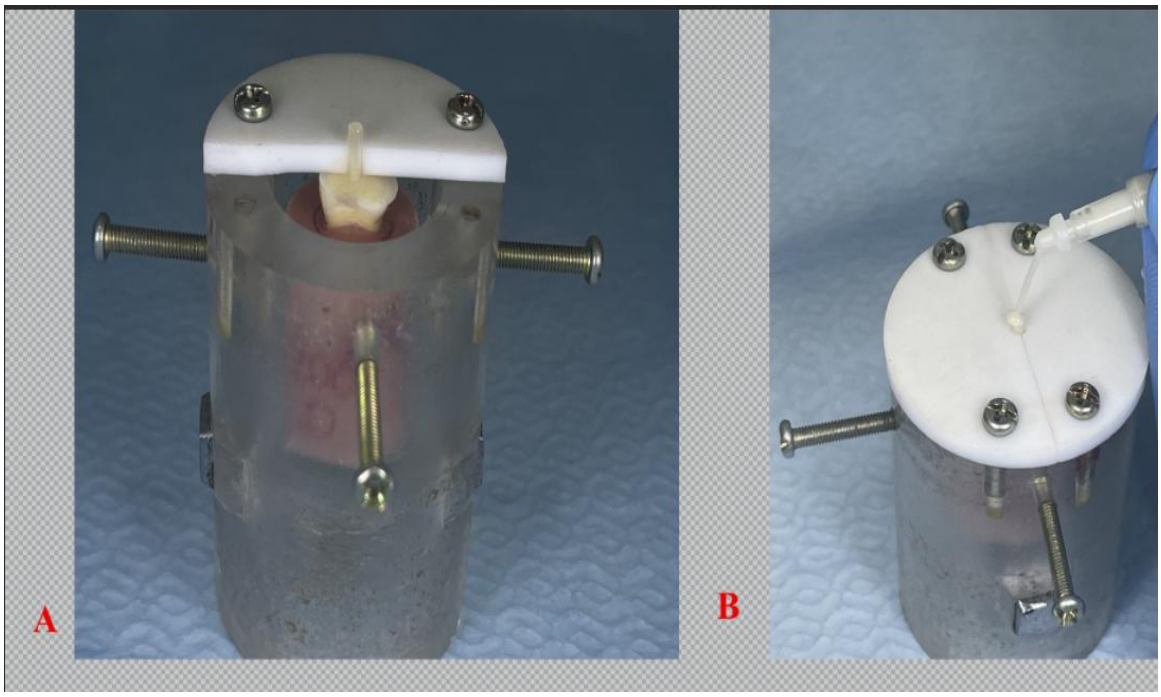

Figure 1: A, The Teflon mold used for cement sample creation. B, insertion of cement through the microtube

The cement was injected as directed by the manufacturer (mixing time: 10 seconds; dispense ratio of the cement (automatic): 1.4:1; working time: 2 min. 30 s; setting time: 5 min. 30 s) (1) . To minimize the amount of air bubbles that might be incorporated into the cement, a red Endo Activator tip (25/0.04, Dentsply Tulsa Dental, USA) was inserted into the cement and activated for 5 minutes at 10,000 cycles per minute (167 Hz) (24). Once set the four cover screws and cylinder fasteners were loosened to remove the specimens from the mold.

All samples were then kept at 37°C in an incubator using deionized distilled water for 24 hours. After storage, a vertical cut was made in each polyethylene microtube with a #11 surgical blade, leaving undamaged cement microcylinders attached to the dentin surface (25). The bonded specimens were examined under 5X magnification loupes (EyeMag Smart, ZEISS, Germany) under LED illumination to detect any air pockets or visible defects; no defective specimens were identified, and all were deemed suitable for testing. Excess cement flash beneath the tube rim was carefully removed using a surgical blade (1) .

2.6 Micro-shear bond strength testing

Cylindrical cement specimens on substrate discs are observed using the micro shear bond test, which also applies a load using a blade to a general testing apparatus. The samples μSBSs were prepared on a matrix which was attached to the universal testing machine (Laryee, China). The shear force was applied to each specimen with the custom-made notched chisel attached to the testing machine, using a 5-kN load cell, at crosshead speed of 0.5 mm/min. The thickness of the central groove of the chisel was 1.2 mm. Up to failure, the load was applied perpendicular to the bonded interface direction. (26). The same set-up was used to make measurements of the six groups at the same time. The μSBS (in MPa) was computed by dividing the maximal load (in N) by the cross sectional surface area (in mm2) (23). This test was conducted in accordance with ISO/TS 11405: 2015's description (27).

2.7 FE-SEM evaluation of the hybrid layer

The failure modes of the objects under investigation were examined using scanning electron microscopy, or FE-SEM (Thermo Fisher Scientific, Axia ChemiSEM). Following the completion of the micro-shear bond strength test, the bonding surface of the typical failed samples was investigated interfacially using Field Emission Scanning Electron Microscopy (FE-SEM). A low-speed diamond saw with water cooling was used to cut the debonded samples in half mesiodistally in order to evaluate the integrity of the hybrid layer and resin tag creation. After then, the divided sections underwent a sequence of dehydration phases with progressively higher ethanol concentrations. After that, the samples were put on aluminium stubs, sputtered with a thin coating of gold-palladium, and examined using a FE-SEM with an acceleration voltage of 5–10 kV. The dentin-cement interface was evaluated at various magnifications to evaluate the hybrid layer's shape, the existence and quality of resin tags, and adhesive failure. (28).

2.8 Failure mode study

The failure modes of the specimens were investigated using scanning electron microscopy (SEM). The SEM is an Axia ChemiSEM from Waltham, Massachusetts, USA-based Thermo Fisher Scientific. First, the samples were dried. After being coated with 45 mA of gold nanoparticles for two minutes, they were analysed using SEM at an operating voltage of 30 kV. A working distance of 10–15.1 mm (x80) was employed. Sticky, mixed largely cohesive, and mixed mostly adhesive failure modes were discovered in the substrates (29)

3. SAMPLE SIZE CALCULATION AND STATISTICAL ANALYSIS

The G*Power software (version 3.1.9.7) was used to undertake an a priori analysis of power in order to determine the minimal sample size. According to the analysis, a one-way ANOVA comparing three main groups for an alpha (α) of 0.05, a power (1-β) of 0.80, and a medium effect size (f = 0.25) required a total sample size of 60 (30) specimens. As a result, sixty undamaged human maxillary premolars were chosen and split into three main experimental groups, each consisting of twenty specimens: Group I (PAA), Group II (GA), and Group III (PAA+GA). Subgroups A (immediate) (n = 10 per subgroup) and B (delayed) (n = 10 per subgroup) were then created from the main groups based on how long they had been ageing before testing.

The data was analysed using IBM SPSS software version 20.0 (Armonk, NY: IBM Corp.). The qualitative data was presented using percentages and frequencies. The Shapiro-Wilk test was used to ascertain whether the data had a normal distribution. Means and standard deviations were used to display quantitative data. The Chi-square test (or, if needed, Fisher's exact test) was used to analyse the differences in categorical variables between groups. One-way ANOVA was used for quantitative normally distributed data, and Tukey's test was used for pairwise comparisons. A paired t-test was used to compare the immediate (24-hour) and delayed (3-month) μSBS within each main group. The significance level of $p < 0.05$ was reached.

4. RESULTS

In general, the PAA+GA group consistently exhibited the highest micro-shear bond strength across both time points, with values reaching 14.19 MPa (±2.07) at 24 hours and 13.97 MPa (±2.07) at 3 months. This outperformed both PAA (13.18 MPa (±1.37) at 24 hrs; 10.18 MPa (±1.67) at 3 months) and GA (9.38 MPa (±1.44) at 24 hrs; 7.64 Mpa (±1.64) at 3 months). Significant differences were observed at 24 hours, PAA+GA was significantly higher than GA ($p_3<0.001^*$), while PAA also showed superiority over GA($p_1<0.001^*$). No significant difference was found between PAA+GA and PAA ($p_2= 0.372$). At 3 months, PAA+GA remained
significantly higher than both GA ($p_3< 0.001^*$) and PAA ($p_2<0.001^*$), As seen in Table 2 and Figure 2, the study's findings really show that PAA performs better than GA (p1 = 0.011*).

Table (2): Descriptive and comparative statistics of μSBS for FujiCem Evolve at 24 hrs and 3 months

FujiCem Evolve		Micro-Shear bond strength (MPa)			p
		PAA (n=10)	GA (n=10)	PAA+GA (n=10)	
24 hrs.	Mean (± SD)	13.18aA (1.37)	9.38bA (1.44)	14.19aA (2.07)	**<0.001***
	Significance between groups	p1<0.001*, p2=0.372, p3<0.001*			
3 months	Mean (± SD)	10.18aB (1.67)	7.64bB (1.64)	13.97cA (2.07)	**<0.001***
	Significance between groups	p1=0.011*, p2<0.001*, p3<0.001*			
p0		0.002*	0.032*	0.805	

p: p value for **Post Hoc Test (Tukey)** for comparing the three studied groups
p1: p value for **Post Hoc Test (Tukey)** for comparing **PAA** and **GA**
p2: p value for **Post Hoc Test (Tukey)** for comparing between **PAA** and **PAA+GA**
p3: p value for **Post Hoc Test (Tukey)** for comparing between **GA** and **PAA+GA**
p0: p value for **Paired t-test** for comparing between **24 hrs**. and **3 months** in each group
*: Statistically significant at $p \leq 0.05$
-Means in the same **raw** with **small common letters** are not significant (i.e. Means with **Different letters** are significant)
- Means in the same **column** with **capital common letters** are not significant (i.e. Means with **Different letters** are significant)

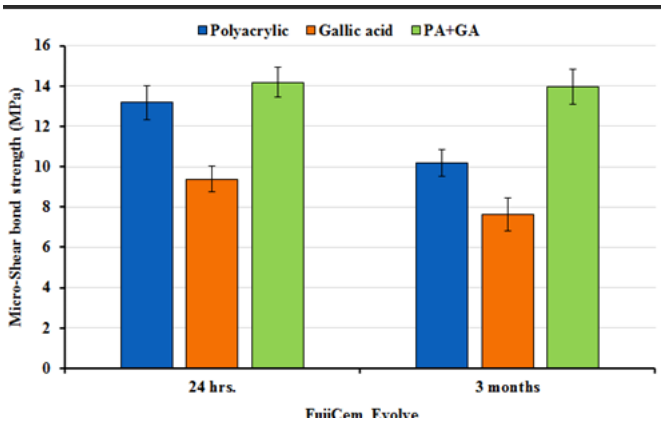

Figure 2 : Bar chart comparing the three studied groups according of FujiCem Evolve at 24 hrs and 3 months

Table 2 and Figure 2 showed that in PAA Group at 24 hours, the bond strength was 13.18 ± 1.37 MPa which significantly decreased to 10.18 ± 1.67 MPa after 3 months (p=0.002*). in GA Group decreased significantly from 9.38 ± 1.44 MPa at 24 hours to 7.64 ± 1.64 MPa at 3 months (p=0.032*). PAA+GA Combination showing 14.19 ± 2.07 MPa at 24 hours and 13.97 ± 2.07 MPa at 3 months not significantly (p = 0.805).

5. FAILURE MODE ANALYSIS

Table 3 and Figure 3 summarise every experimental group at 24 hours and 3 months. Adhesive failure was the most frequent mode of failure across all groups and time periods. For the initial 24-hour evaluation, there was a significant difference (p=0.037) in the failure mode distribution between the groups. The GA group demonstrated solely adhesive failures (100%), a rate markedly superior to that of the PAA (40%) and PAA+GA (40%) groups. Cohesive failures were infrequent and confined to the PAA (20%) and PAA+GA (20%) groups. Likewise, mixed failures occurred in the PAA (40%) and PAA+GA (40%) groups but were completely absent in the GAgroup. Three months (delayed) assessment and periodic differences showed the distributions of failure modes changed, resulting in the total inter-group differences which were statistically significant. The GA group exhibited no alterations, maintaining a steadfast 100% adhesive failure rate without any cohesive or mixed failures.

Significant changes were noted among the other groups over time. Within the PAA group, the incidence of adhesive failures jumped from 40% to 70%; however, this variation was statistically significant relative to its 24-hour baseline (p=0.025). This was accompanied by a significant reduction in cohesive failures (from 20% to 10%) and mixed failures (from 40% to 20%). The PAA+GA group exhibited non-significant differences compared to baseline period with a decrease in adhesive failure rate of from (40%) to (30%). Cohesive failures were increased to be (30%). However, mixed failures are unchanged (40%). The persistent lack of cohesive and mixed failures in the GA group at both time intervals highlights its failure to create a lasting, cohesive bonding contact with the substrate. The various failure modes observed in the debonded specimens are represented by the SEM photomicrographs shown in Figure 4.

Table (3): Comparison between the three studied groups according to Failure mode in 24 hrs and 3 months

Failure mode	Polyacrylic (n=10)	Gallic acid (n=10)	PAA+GA (n=10)	χ^2	^{FE}p
Evolve 24 hr					
Adhesive	4 (40%)	10 (100%)	4 (40%)	8.396*	0.037*
Cohesive	2 (20%)	0 (0%)	2 (20%)		
Mixed	4 (40%)	0 (0%)	4 (40%)		
Evolve 3 months					
Adhesive	7 (70%)	10 (100%)	3 (30%)	6.666*	0.024*
Cohesive	1 (10.0%)	0 (0%)	3 (30%)		
Mixed	2 (20%)	0 (0%)	4 (40%)		

* Statistically significant at **p ≤ 0.05**

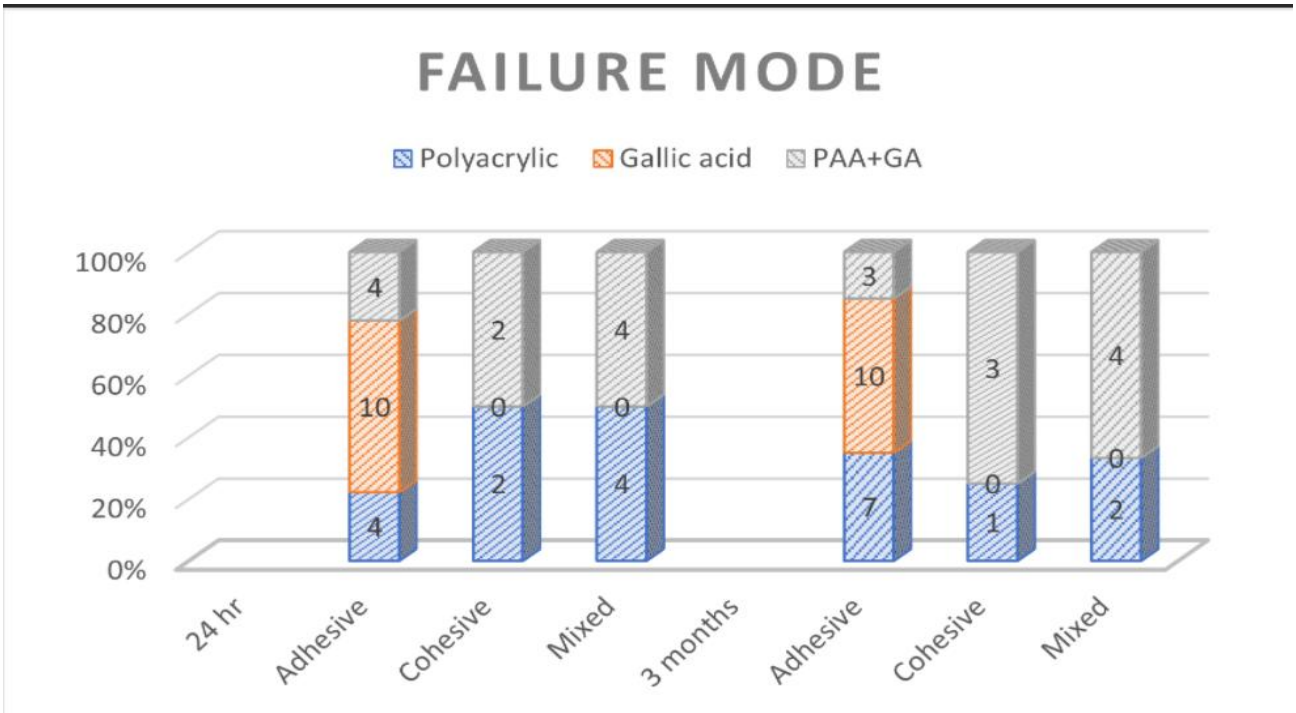

Figure 3: Comparison between the three studied groups according to Failure mode in 24 hrs and 3 months

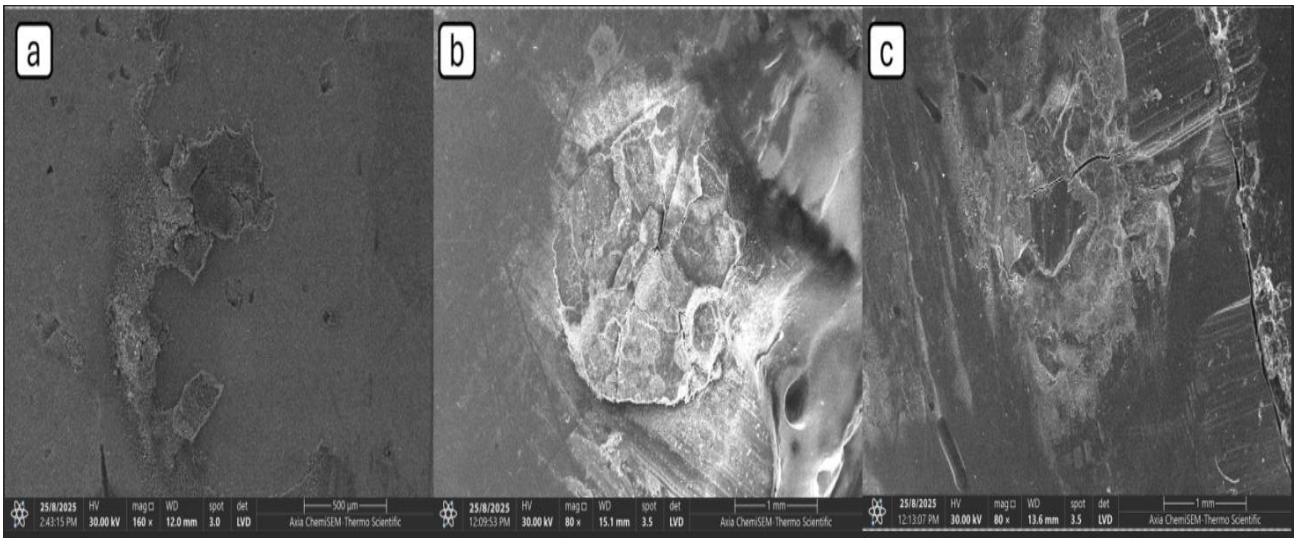

Figure 4: showing different failure modes. a: mixed failure. b, cohesive failure. c, adhesive failure.

6. HYBRID LAYER FINDINGS BY FE-SEM

The FE-SEM analysis of the debonded interfaces revealed unique morphology for each group. The PAA+GA group demonstrated a distinct hybrid layer characterized by numerous extended resin tags infiltrating the dentinal tubules and irregularities. Conversely, the PAA group exhibited a discernible hybrid layer, however with significantly shorter and fewer resin tags. The GA group had the least interaction, marked by a thin hybrid layer and little, sparse production of resin tags as shown in figure 5.

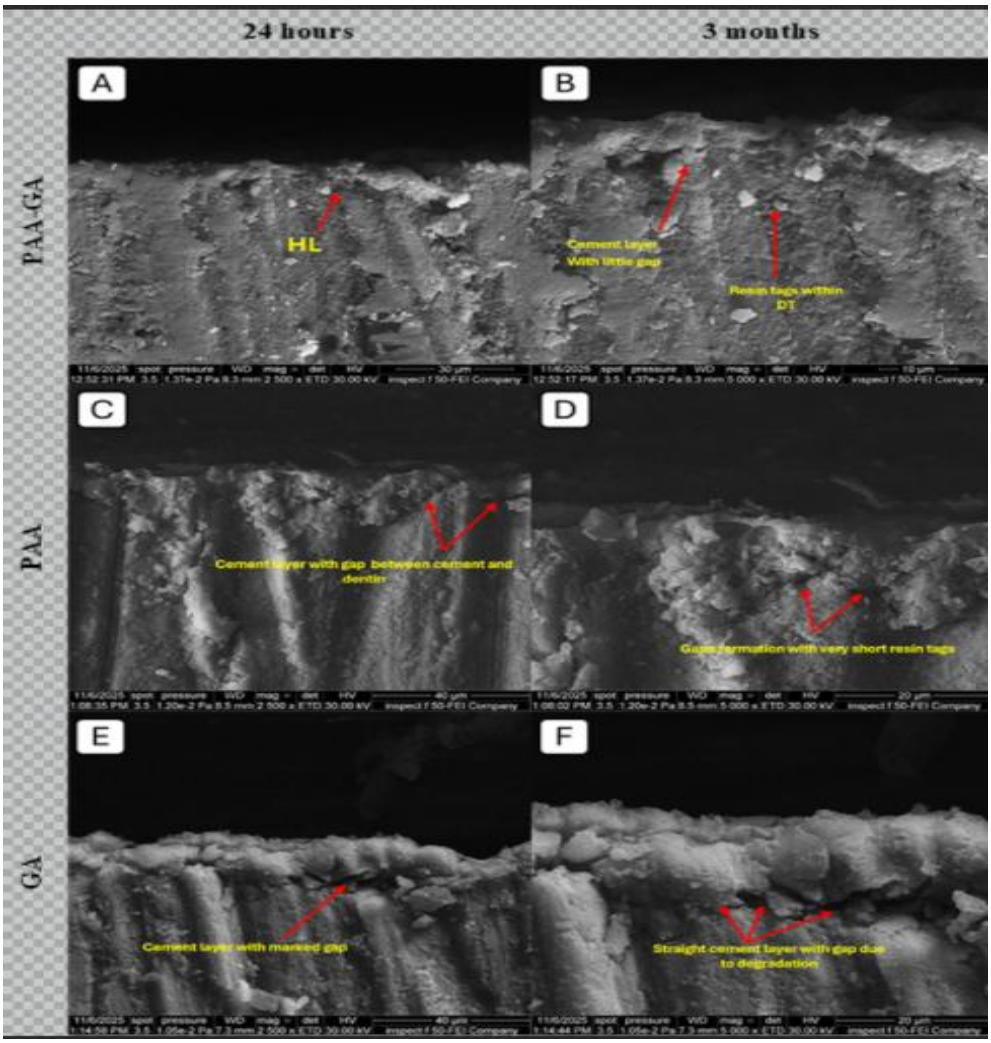

Figure 5: FE-SEM representative photomicrographs demonstrating the ultrastructural features and aging of the resin-dentin interface. Panels (A–B) illustrate the enhanced performance of the PAA+GA group, distinguished by a clearly defined hybrid layer (HL), extensive and durable resin tags (RT) reflecting close adaptation of cement layer to dentin, strong micromechanical interlocking, and stable chemical adhesion to dentin collagen, with no evidence of degradation following 3 months of aging. In contrast, the PAA group (C-D) although showing good adhesion it also exhibits a compromised interface characterized by shorter resin tags, gap formation, and clear deterioration of the hybrid layer and resinous materials. The GA group (E-F) demonstrates a superficial, linear, and weakly delineated hybrid layer resulting from inadequate micromechanical retention (weak acid), with marked gaps formation, emphasizing the essential importance of both micromechanical and chemical bonding for achieving durable adhesion.

7. DISCUSSION

In the present study, the concentrations and application protocols for the conditioners were selected based on preliminary studies and existing literature to balance efficacy with biocompatibility. A 15% PAA solution is established as an effective conditioner for pretreatment of dentin surface for GICs, providing optimal smear layer removal without excessive demineralization that could compromise the dentin collagen network (21, 31). GA (1%) was chosen as it represents a concentration demonstrated to provide significant antioxidant and collagen cross-linking activity without excessively inhibiting the setting reaction of the adjacent RMGIC; the one minute application time for GA was adapted from protocols of other bioactive agents to allow sufficient interaction with dentin substrate. This level of concentration was chosen because it has been shown in the literature to be the most effective in improving the endurance of resin-dentin bonds by striking a compromise between allowing enough resin monomer entry and adequate collagen crosslinking and antioxidant activity (18).

In this in vitro investigation, the dentin surface pretreatment with polyacrylic acid (15%) and galic acid (1%) was assessed for the μSBS of glass ionomer cement (Fuji CEM evolve). The first null hypothesis has been rejected because, according to the results, null hypothesis 2 acknowledges that the micro-shear bond strength of RMGIC luting cement in mixed technique (PAA + GA) significantly maintains the micro-shear bond strength of RMGIC luting cement after three months of storage in distilled water. The third null hypothesis has been accepted since the analysis of failure modes supported the

mechanical results. The GA group's high frequency of adhesive failures and the PAA and PAA+GA groups' higher incidence of mixed failures demonstrated the relative durability and integrity of the adhesive interface created by each treatment.

The dentin and resin-based materials bond strength is necessary for the dental restorations long-run outcomes. Over time, however, the hybrid layer, which forms at the interface between dentin and adhesive materials, can degrade due to factors such as hydrolytic breakdown and oxidative stress. This degradation weakens the bond and compromises the durability of restorations.

Antioxidant GA have been explored as potential agent to protect the dentin surface by neutralizing reactive oxygen species and stabilizing the collagen matrix, thereby potentially improving bond strength and longevity. Therefore, this study estimated the GA, PAA, and their combination effect on the μSBSs of FujiCEM Evolve GIC.

In this study, it was found that the GA and PAA combination significantly enhanced the μSBS after both 24 hrs and 3 months compared to the control. This finding suggests that the synergistic effect of GA – which provides an antioxidant effect, collagen-stabilizing ability and weak acidity – combined with the moderately acidic PAA, contributes to smear layer (SL) removal, as well as improved micromechanical interlocking and chemical bonding. GA, a natural polyphenol, is accepted to boost collagen cross-linking and inhibit MMPs, so stabilizing the dentin substrate over time (32). PAA's and GA capacity to partially remove the SL and expose hydroxyapatite for ionic bonding; and the acidic nature of both agents' promoting surface demineralization to facilitate micromechanical interlocking. Our findings corroborate those of Hardan et al., who found that collagen cross-linkers improve bond longevity by increasing the integrity of the hybrid layer (33). After three months, the GA+PAA group's results support the theory because the treatment significantly improved the μSBS's performance in comparison to the other groups. Additionally, GA has been shown to be anti-inflammatory because it reduces the expression of inflammatory cytokines including IL-6 and interleukin-1β (IL-1β), which are often increased during carious or inflamed dentin. By downregulating these pro-inflammatory mediators, GA may contribute to a more biologically favourable bonding environment, minimizing postoperative sensitivity and promoting pulp healing. This dual bifunctional role—mechanical enhancement and anti-inflammatory action—makes GA a promising dentin surface pretreatment agent (32).

There are two types of bonding mechanisms between GICs and dentin: chemical contact and micro-mechanical interlocking (34, 35). The lifetime of the RMGIC repair depends on the quality of the bond strength, which is created on the surface during cavity preparation when rotary instruments are used (36, 37). The presence of a layer that inhibits the chemical bonding of RMGIC to the intratubular dentin and limits any micromechanical interlocking to the collagen may impair the bonding capability of RMGIC if the residue is not removed from the dentin surface prior to application (38, 39). Conditioners and adhesives may dissolve or eliminate the residue layer to obtain optimal adhesion to the tooth surface, as it does not adhere securely to the surface of tooth (40). Nevertheless, RMGICs may bind to the smear layer in a variety of methods. The PAA in RMGIC can be a mild dissolvent for the residue layer which acts as a moderate self-conditioner. Chemical bonding of calcium ions with the polyalkenoic acid molecules in the RMGIC may be facilitated in the SL calcium ions. The cavity conditioner is a PAA conditioner that demineralizes the dentin and partially eliminates the surface layer without severing dentin tubules. The enhanced surface area and micro porosities, resulting from this procedure, will increase the bond strength of the RMGIC (38, 39, 41). Similarly, the extra micromechanical retention is caused by the interaction of Ca ions between the exposed collagen and the hydroxyapatite linked to the COOH group of polyalkenoic acid (38). Thus, a gel phase settled over the hybrid layer of PAA-pretreated dentin surface as an intermediate formation (35, 42). The conditioner PAA carboxyl groups and hydroxyapatite calcium chemical reaction are thought to be the cause of the calcium-polycarboxylate salt accumulation that is represented by the gel phase (42). Therefore, in the RMGIC bonding mechanism, the interaction may be more important than the interlocking in the micro-mechanical bind to dentin (caused by hybridization). GICs adhere to dentin by two bonding mechanisms: chemical contact and micro-mechanical interlocking (34, 35). The quality of the bond strength is crucial for the clinical longevity of the RMGIC restorations since a film layer will be generated on the surface during cavity preparation due to the employment of rotating devices (36).

Conversely, it has been asserted that the cavity conditioner utilization was insignificant in enhancing the RMGIC bond strength to dentin without a SL (35). All the experimental group flat dentin surfaces. When compared to the control group, the RMGIC's dentin bond strength increased as a result of the Ugurlu study, which established a standard SL (43), as covered in previous research (35, 39, 41). Additionally, the removal of the smear layer, which enhances dentin permeability, may supply an additional water source for the GIC acid-base setting reaction (44). Therefore, the maturation of GIC at the interface could facilitate the RMGIC adhesion to dentin, thereby increasing its resistance to degradation over time (45). Last but not least, a number of variables, including PH, viscosity, and chemical makeup, may affect the resin cement's capacity to adhere to dentin. It has been hypothesized that the bond between this cement and dental structure could be adversely affected if the pH is maintained for an extended period, despite the fact that a low pH is necessary for adequate dentin debridement (38, 46). In such cases, where the pH is low enough, the smear layer may be partially removed and the RMGIC matrix can penetrate the dentin superficially, creating a zone of interdiffusion between the cement and the matrix. The Cavity Conditioner's pH is 1.2 (47).

These findings are consistent with those of Andrade et al., who showed that adding natural plant extracts to GICs enhances mechanical characteristics only up to a threshold concentration, after which deterioration takes place (48).

In the present study, it was found that at 24 hours, adhesive failures dominated the GA group (100%), significantly more than the PAA (40%) and PAA+GA (40%) groups ($p = 0.037$), reflecting its weaker interfacial adhesion. The incidence of cohesive and mixed failures was higher for the PAA+GA group, especially at 3 months (40% and 30%, respectively), which is important to note, indicating a more integrated bond between cement and dentin that was resistant to ageing degradation. The observed transition toward mixed failure in the PAA+GA group, which became more pronounced after 3 months, is a critical indicator of a more durable and integrated bond. This implies that the combined treatment established an interface in which the failure path was not exclusively located at the adhesive junction, but occasionally extended to the underlying dentin or the overlying cement. This is likely the result of PAA facilitating micromechanical interlocking through partial demineralisation, while GA simultaneously stabilises the exposed collagen matrix against hydrolytic degradation. The transition to solely adhesive failure observed in the other groups is frequently caused by the weakening of the hybrid layer over time, which is prevented by this synergistic stabilisation approach.

The visual evidence provided by the FE-SEM analysis of the debonded interfaces is persuasive and directly supports the μSBS and failure mode results. The PAA+GA exhibited an interfacial morphology that was distinguished by a robust hybrid layer, numerous, distinctly defined resin bands that penetrated the dentinal tubules, and micro-irregularities.

This observation signifies efficient micromechanical interlocking, resulting from the synergistic demineralization by PAA and the mild acidity of GA. Furthermore, the integrity of this interface following three months of water ageing shows that GA's collagen-stabilizing effect, through its antioxidant and crosslinking properties, successfully prevented the hybrid layer's enzymatic and hydrolytic degradation, preserving both the micromechanical and chemical bonds. In contrast, the FSEM pictures of the PAA group displayed only minor and partially destroyed resin tags, indicating that the bond strength depended predominantly on micromechanical interlocking, which diminished with time without a collagen-stabilizing agent. The GA group demonstrated the weakest interfacial connection, characterized by minimal cement penetration and a significant lack of robust resin tag production, correlating with its limited demineralizing ability and elucidating the primarily adhesive failure mode along with the lowest μSBS values. The FS-EM results unequivocally indicate that the combination of PAA and GA produces a superior and more degradation-resistant adhesive interface as shown in figure 5.

The failure modes of RMGIC were reported to be variable in previous studies. (39, 49, 50). It has been suggested that adhesive failure is primarily predictable during the initial stages of development; however, the frequency of cohesive failures may rise as the product ages (34). The prevalence of cohesive and mixed failure types (51) may be caused by voids created as a result of entrapment of air in the material during hand blending. By enhancing the chemical adhesion of RMGIC through the use of various functional monomers in universal adhesives, the dentin bond strength is improved, as indicated by previous research (41, 49, 50, 52). Because RMGIC contains HEMA and other resins, the adhesives may react with it (38, 51). This increase in bond strength is thought to be caused by dentin's improved wettability and ease of monomer infiltration, which enhances the quality of the hybrid layer—the primary source of bonding (38, 47).

It is well known that they are made of resin composite technology, even though there is no evidence in the literature that the combination of PAA and GA would alter the mechanical properties of the self-adhesive RC. Their adhesive properties also contribute to their effectiveness, which is greatly affected by the resistance to masticatory forces (53). Vidal et al., in their research, found that GA was able to cross-link the collagen structure, reducing the rate at which it is biodegraded in dentin. Moreover, GA derivatives showed to improve the dentin matrix mechanical properties during the investigation (54).

The majority of the recent research investigations on the hybrid layer integrity improvement and longevity through the GA derivatives used was carried out by directly adding the normal cross-linkers to the adhesive bonding agents. Du et al. (55) found that adding the GA derivative epigallocatechin-3-gallate (EGCG) to the dental glue had a beneficial therapeutic impact, such as increased microtensile bond strength and antibacterial activity. Neri et al. performed another study that did not show the EGCG to compromise either the conversion degree or flexure strength of a self-etch bonding agent (56). Hu et al. (24) examined the GIC's physical properties with the addition of EGCG and discovered that, as compared to the control groups, the flexural strength and surface hardness had improved.

Nevertheless, the mechanical characteristics of the assessed RCs in the Oguz Ahmed study were poor and connected with the GA addition (57). This could be because phenolic chemicals have an inhibiting effect on methacrylate monomer free-radical polymerisation (58). Nevertheless, additional pertinent research should be conducted to substantiate the conclusions of this investigation. Additionally, additional investigations may evaluate various aging conditions.

This in vitro study offers important insights into the possible synergistic effect of the PAA and GA; however, when analysing the data, it is important to keep in mind a few constraints.

1. Utilising in vitro design, which is done in a controlled laboratory setting, does not fully mimic the complicated oral environment. The occlusal load, thermal cycling and enzymatic activity in saliva were not simulated. Future studies using thermocycling and mechanical fatigue would give a more clinically relevant evaluation of bond durability.
2. Dentin substrate variability, that despite efforts to standardize the dentin surface, natural variations in tubular density from and mineralization between teeth from different donors could introduce inherent variability in bond strength measurements. The use of teeth from a relatively young age group (14-30 years) may also not represent the dentin older patients.

3. Single concentration evaluation of PAA (15%) and GA (1%), the optimal synergistic effect might exist at different concentrations or application times not explored in this study. A dose-response analysis would be necessary to identify the most effective clinical protocol.
4. The proposed mechanism of action, particularly the chemical interaction between GA and the RMGIC components or the dentin matrix, remains speculative without direct analysis evidence. Because of the nature of these interactions, methods such as Raman spectroscopy and/or Fourier-Transform Infrared Spectroscopy (FTIR) would be necessary.

CONCLUSION

When compared to separate treatments, the FujiCem Evolve dentin bond (PAA+GA) exhibited significantly better dentin bond strength and endurance. Polyacrylic alone initially produced a reasonable binding strength, but over time, it showed a noticeable drop. The GA treatment continuously showed poor bonding performance and substantial adhesive failure rates. On the other hand, the PAA+GA group showed a more favourable distribution of failure modes and had a steady bond strength over a three-month period, indicating that it had better long-term stability. These results imply that polyacrylic and GA pretreatment have a synergistic effect, providing a viable method for enhancing the clinical performance of RMGICs.

Statements and Declarations

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Competing Interest:

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Ethical approval:

This study involving human subjects was carried out in compliance with the World Medical Association's Code of Ethics (Declaration of Helsinki). The University of Diyala's Institutional Review Board (IRB) authorized the research after reviewing its protocol (Approval No: *2025RJK966* / Date: 22 Oct 2025).

Informed Consent

The extracted teeth used in this study were obtained from (my own clinic and the specialized dental health center in B'aquba/ Diyala Governorate). The original donors provided informed consent for the use of their extracted teeth in research.

Data Availability: The corresponding author can provide raw data upon reasonable request.

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