Original Research
Dental Materials

Effect of gallic acid addition on some mechanical properties of self-adhesive resin cements

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Abstract: Self-adhesive resin cements (RCs) activate matrix metalloproteinase (MMP) and cathepsin-related collagen degradation, and gallic acid (GA) inhibits the activity of both MMPs and cysteine cathepsins. The purpose of this study was to evaluate the setting time, biaxial flexural strength, and Vickers hardness of self-adhesive RCs after the addition of two different concentrations of GA. RelyX U200 (3M ESPE) and Panavia SA (Kuraray) were modified with 0.5 and 1 wt% GA. The setting time of five samples in each RC group was assessed using a thermocouple apparatus as described in the ISO 4049 test. Biaxial flexure strength was measured using a universal testing machine until failure. Vickers hardness was measured with three randomized indentations on the surface of each resin disc. RCs without GA were used as control. Data were analyzed using a one-way analysis of variance and Tukey's HSD test ($\alpha = 0.05$). The setting times ranged from 2.4 to 4.6 min for RelyX and from 4.9 to 6.0 min for Panavia. The biaxial flexure strength ranged from 76.5 to 109.7 MPa for RelyX and from 73.3 to 108.2 MPa for Panavia. Vickers hardness values ranged from 41.6 to 58.6 for RelyX and 27.2 to 33.6 for Panavia. The addition of 0.5 and 1 wt% GA to improve durability of resin-dentin bonds had no adverse effects on setting time, whereas the biaxial flexure strength and Vickers hardness values for the tested materials were significantly reduced.

Keywords: Resin Cements; Gallic Acid; Hardness.

Introduction

Ceramic restorations are important for the current aesthetically driven dental practice. Apart from inlays and onlays, which are fabricated after minimal reduction of tooth structure, all-ceramic full crowns also require adhesive cementation.[1] Therefore, clinicians have increased the use of self-adhesive RCs based on filled polymers, partially because of the simplicity of their application which does not require separate etching and bonding procedures. In the recent years, various proprietary products have been introduced in the market owing to the ease of application of these luting cements.[2]

The success of adhesively bonded restorations depends on the bond strength between the resin–restoration and resin–dentin interface. The latter interface, also known as "hybrid layer," is created by the infiltration

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and ensuing polymerisation of resin monomers within the demineralized microporous zone of dentin matrix.[3] Thus, to prolong the survival of adhesively bonded restorations, an optimal hybrid layer is required for resisting several degradation mechanisms.[4] Among the degradation patterns of the resin–dentin interface, degradation of exposed collagen fibrils by endogenous dentin proteases such as matrix metalloproteinases (MMPs) and cysteine cathepsins (CCs) occur as a result of acid-etching procedures. Such a degradation pattern could be triggered by etch-and-rinse or self-etch adhesive systems as well as the simultaneous demineralization of the dentin layer by acidic monomers included in self-adhesive RCs.[4,5,6,7]

In several studies, plant extracts which are rich in proanthocyanidin (PAC) have been shown to be efficient natural cross-linking agents for improving biomechanical properties, integrity and stability of the hybrid layer of resin–dentin bonds.[8,9] Gallic acid (GA) is a type of water-soluble phenolic acid, also known as 3,4,5-trihydroxybenzoic acid, as well as one of the main polyphenols extracted from green tea leaves and grape seeds; GA also has good antioxidant properties.[10,11,12,13] GA moieties, such as catechin gallates and gallocatechins, are present in both of the different "gallo" motifs of oligomeric PACs (OPACs),[14] and they were shown to inhibit the activity of both MMPs and CCs.[15]

Addition of active ingredients in proprietary materials could result in enhanced antimicrobial activity[16] or improved protection against enzymatic degradation.[17,18] It is essential to evaluate the currently used adhesive resin cements for changes in their mechanical properties following modifications by the incorporation of active ingredients. Giannini et al.[19] reported that the tests for biaxial flexure strength provided reliable results since the maximum tensile stresses occur within the central loading area of the disc-shaped specimens. The flexural strength data provides valuable information regarding the ability of the cement to resist forces and thereby absorb energy without any permanent deformation.[20] Vickers hardness is a non-destructive testing method suitable for measuring the hardness of dental cements,[21] and provides significant data associated with the wear resistance of dental cements.[22] While microhardness and flexure strength correlates with the quality of polymerisation of resin-based materials, setting time gives useful information about the changes in polymerisation reaction kinetics depending on the various ingredients added in self-adhesive RCs.[23,24,25] To the best of the author's knowledge, the effects of GA addition on the mechanical properties of self-adhesive RCs has not been studied. Therefore, the aim of this study was to evaluate Vickers hardness (VH), setting time $(t)_s$ and biaxial flexural strength of self-adhesive RCs after the addition of gallic acid at two different concentrations and to analyse the effects of gallic acid on some mechanical properties of these materials. The hypothesis tested was that the addition of GA into self-adhesive RCs did not adversely affect their physical properties.

Methodology

The following self-adhesive RCs were tested in this study: Panavia SA (Kuraray Noritake Dental Inc. Okayama, Japan) and RelyX U200 (3 M ESPE, Neuss, Germany) (Table); 1% and 0.5% GA were prepared from the stock solution of 20% GA in ethanol. The final concentration of gallic acid in cement was calculated as weight to prepare mixtures containing two different concentrations of GA: 0.5 wt% and 1 wt%. The solutions were manually incorporated into the cements during mixing. Materials without GA addition served as positive control, whereas materials with ethanol addition served as negative control.

Setting time measurements

Setting time of the tested materials was assessed using a thermocouple apparatus, as described in the ISO 4049 test.[17] The apparatus consisted of a polyamide block measuring 6 mm in diameter, with a locating part measuring 4 mm in diameter and 2 mm in height. A 6-mm long and 1-mm thick high-density polyethylene tubing fitting into the locating part was used to form the specimen well. A T-type thermocouple wire surrounded by steel tubing was located inside the polyamide block [ISO 4049:2009 (E)]. To facilitate the removal of samples after testing, the thermocouple junction had a conical

Table. Materials used in the study. Bis-GMa, Bisphenol A-diglycidylmethacrylate; MDP Monomer, 10-Methacryloyloxydecyl dihydrogen phosphate; TEGDMA, triethylene glicol dimethacrylate.

Name	Type	Manufacturer	Total filler content	Monomers	Shade	Lot numbers
RelyX U200™	Dual-curing self-adhesive resin cement paste/paste automix	3M ESPE, Neuss, Germany	43%	Paste A: Bis-GMA, TEGDMA, photoinitiators, inorganic particles of silica and zirconia (68% by weight), and pigments. Paste B: TEGDMA, Bis-GMA, inorganic particles of silica and zirconia (67% by weight), benzoyl peroxide.	A2	583827
Panavia™ SA	Dual-curing self-adhesive resin cement paste/paste automix	Kuraray, Europe GmbH, Hattersheim am Main, Germany	40%	Paste A: 10-MDP, Bis-GMA, TEGDMA, Hydrophobic aromatic dimethacrylate, Silanated barium glass filler, Silanated colloidal silica, Dl-Camphorquinone, Benzoyl peroxide, Initiators Paste B: Bis-GMA, Hydrophobic aromatic dimethacrylate, Hydrophobic aliphatic dimethacrylate, Silanated barium glass filler, Silanated colloidal silica, Surface treated sodium fluoride, Accelerators, Pigments	A2	920062

solder tip which protruded 1 mm into the base of the specimen well. Five samples for RelyX and Panavia including 0.5 wt% and 1 wt% GA, no-GA as the positive control and ethanol as the negative control were poured and mixed into the specimen well. The time elapsed before the mixing of the adhesive resin cements was recorded. Before and during the test, the apparatus was stored in an incubator at 37°C. An increase in temperature was recorded at a sampling rate of 10 Hz using a thermocouple data logger with a built-in cold junction compensation (TC-08, Pico Technologies Ltd., Cambridgeshire, UK). Measurement was continued until a plateau was reached at the maximum temperature. The plateau was extended backwards to meet the straight line of increase in temperature. Time at the intersection of the both lines was recorded as the setting time, $(t)_s$.

Biaxial flexural strength measurements

Disc-shaped specimens (0.5 mm thick and 7.0 mm diameter) were fabricated in a polyethylene mould after 40-s light exposure on both sides with a halogen light curing unit (XL 3000; 3 M, ESPE, St Paul, MN) with an output intensity of 600 mW/cm^2. Before the mechanical tests were applied, specimens were stored at 37°C for 24 h in a dark container. Ten specimens of both materials at two different concentrations (0.5 wt% and 1 wt%) of GA were tested (n=10). Therefore, a total of 80 specimens were fabricated including the negative and positive control groups. Each specimen was placed into a custom-made testing jig and was tested in biaxial flexure on a universal testing machine (Model LR30KPlus; Lloyd Instruments Ltd., Fareham, UK) at 1.27 mm/min until failure. The testing jig consisted of a stainless steel plate with a 6 mm diameter recess at the center. The deeper borders of the recess were shaped to support the outer edges of the specimen during testing. The discs were loaded until failure at the center using a 1 mm diameter stainless steel plunger. The maximum load was recorded for each specimen, and the following formula for the biaxial flexural strength (σ) was used:[26]

$$\frac{7P\,(X - Y)}{b^2}$$

Centre tensile stress (Megapascals),

P is the total load causing fracture (Newton),

$X = (1 + v)\, ln(r_2/r_3)^2 + [(1 - v)/2]\, (r_2/r_3)^2$

$Y = (1 + v)\, [1 + ln(r_1/r_3)^2] + [(1 - v)\, (r_1/r_3)^2]$

and b is the specimen thickness at fracture origin (millimetres), in which v is Poisson's ratio (used $v=0.25$), r_1 is the radius of the support circle (millimetres), r_2 is the radius of the loaded area (millimetres) and r_3 is the radius of the specimen (millimetres).

Vickers hardness measurements

Resin discs were prepared in a mould with a thickness of 0.5 mm and a diameter of 7 mm (n=10 for each group). A polyester strip and a glass

slide was placed on the paste and gently pressed to expel the excess material. A halogen curing-unit tip (XL 3000; 3M, ESPE, St Paul, USA) with an output intensity of 600 mW/cm^2 was positioned at 1mm from the adhesive surface during photo polymerisation, and the specimen was light-cured for 40 s. Following storage of the specimen at 37°C for 24 h in a dark container, the Vickers indenter was applied to self-adhesive resin cement disc surfaces at a load of 490.6 mN and with a dwell time of 15 s. Three randomized indentations were made on the bottom surfaces of each resin disc with each indentation separated by ~0.5 mm. Microhardness was measured using a Struers Duramin hardness microscope (Struers, Copenhagen, Denmark) with a 40× objective lens. The diagonal length impressions were measured and Vickers values were converted into microhardness values by the machine. Microhardness was obtained using the following standard formula (ASTM E384, 1991)[27]: $H = 1.854P/d^2$, where H is Vickers hardness in N/mm^2, P is the load in N and d is the length of the diagonals in mm.

Statistical analysis

Shapiro–Wilk test was applied to the test results to confirm normal distribution of the data. One-way analysis of variance (ANOVA) using Tukey's HSD post-hoc analysis (α= 0.05) was used to detect any statistically significant differences among the groups (IBM SPSS Statistics Software version 21, IBM Corporation, Armond, USA).

Results

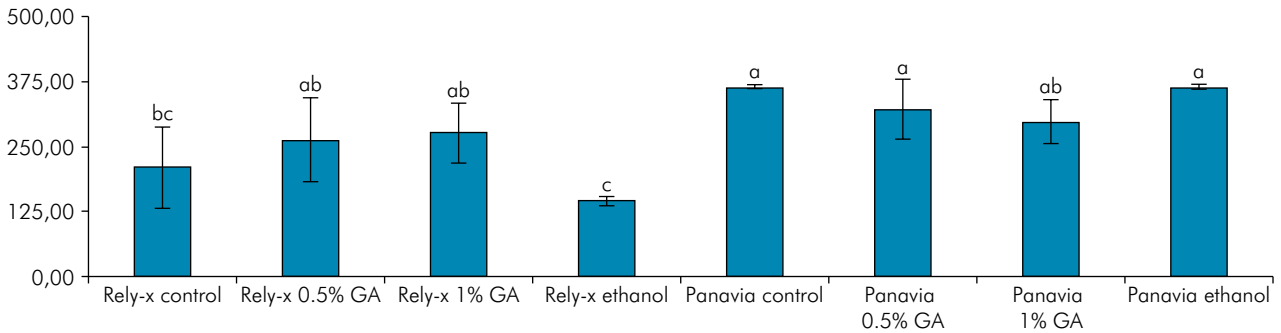
Setting time values ranged from 2.4 to 4.6 min for RelyX and from 4.9 to 6.0 min for Panavia. (Figure 1). Addition of 0.5% and 1% GA did not have any significant effect on the setting time values for both RCs ($p > 0.05$).

Biaxial flexure strength values ranged from 76.5 to 109.7 MPa for RelyX and from 73.3 to 108.2 MPa for Panavia (Figure 2). GA- and ethanol-added groups showed significantly lower biaxial flexure strength than that of the control groups for both RC types ($p < 0.05$).

The results of Vickers hardness are shown in Figure 3. Vickers hardness values ranged from 41.6 to 58.6 for RelyX and from 27.2 to 33.6 for Panavia. Compared with ethanol-added and control groups, the 0.5% and 1% GA-added groups showed significantly lower Vickers hardness values for RelyX ($p < 0.05$). On the other hand, for Panavia samples, addition of 0.5% GA resulted in a significant decrease ($p < 0.05$) in the Vickers hardness values, whereas the addition of 1% GA did not have any significant effect ($p > 0.05$). Overall, although the addition of GA negatively affected the mechanical properties of self-adhesive resin cements, their setting time was not influenced.

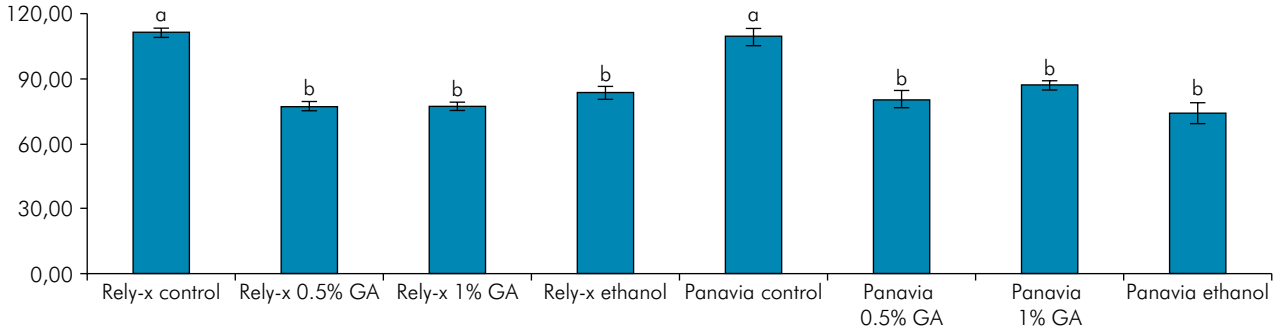
Discussion

Self-adhesive RCs are based on resin composite technology, and their mechanical properties should be carefully considered for the selection of the most



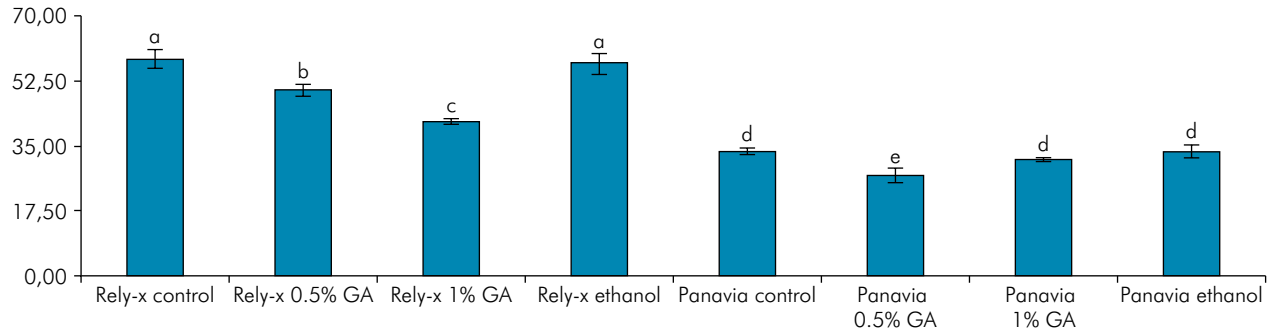
Letters indicate statistically similar groups ($p > 0.05$).

Figure 1. Setting time(s).



Letters indicate statistically similar groups ($p > 0.05$).

Figure 2. Biaxial flexure strength (MPa).



Letters indicate statistically similar groups ($p > 0.05$).

Figure 3. Vickers' hardness.

suitable material in clinical applications. Besides their adhesive properties, their resistance against masticatory forces is an important factor which determines their success.[28] Another important point is the durability of resin–dentin interface created after the application of self-adhesive RCs. Because self-adhesive RCs are chemically composed of strong acidic monomers, they reportedly activated endogenous dentin proteases[7] in a manner similar to the activation of self-etch adhesives.[5]

Owing to their biocompatibility and high dentin bioactivity,[29] the bio-modification of dentin matrices by PACs was previously shown to inhibit the activity of endogenous proteases.[29,30,31,32] In a recent study by Vidal et al.[11] it was observed that GA mimicked the cross-linking mechanism in dentin, which in turn reduced the rate of collagen biodegradation; additionally, the study also revealed that GA derivatives enhanced the mechanical properties of the dentin matrix. Therefore, the present study evaluated some of the mechanical properties of two light-cured self-adhesive RCs after the incorporation of GA present in oligomeric PACs. According to the results, the null hypothesis was partly rejected as the direct addition of GA had significant effects on some of the mechanical properties tested.

In the present study GA was dissolved in ethanol to incorporate the former directly into RCs because its solubility in ethanol is greater than that in water.[33] Therefore, we also prepared ethanol-added negative control groups. Addition of 0.5% and 1% GA prolonged the setting time values for RelyX samples; however, the values were not significant different from those of the control ($p > 0.05$). On the other hand, addition of 0.5% and 1% GA into the Panavia samples decreased the setting time values, which were also not significantly different from those of the control ($p > 0.05$). Besides, ethanol addition into both of the RCs did not have

any adverse effects on the setting time in comparison with the negative control groups. The effects of adding ethanol into the self-adhesive resin cements are not known and warrant further studies.

Resin-bonded partially fixed restorations are frequently subjected to bending forces in the oral environment. Such a loading configuration may also subject the cement to flexure.[34] It was previously reported that biaxial flexure strength data obtained from a three-point flexure strength testing method provided reliable results and also had the advantage of using appropriate size-scale specimens as opposed to the data obtained from three-point bending method.[35] According to the results of the biaxial flexure strength tests, 0.5% GA, 1% GA and ethanol additions produced almost a 30% decrease in the strength of the specimens, which was significantly different from the strength values of the control groups for both RCs.

With the addition of 0.5 and 1% GA into RelyX samples, a significant 20%–30% decrease in hardness was observed in comparison with the hardness values of the control and ethanol-added groups ($p < 0.05$). For Panavia samples, addition of only 0.5% GA had an adverse effect and lowered the hardness values up to 20%, which was statistically significant when compared with the other groups ($p < 0.05$). Ethanol did not have a negative effect on the hardness values in most of the groups. It was reported that microhardness of a specific resin-based material correlates with its degree of conversion during the setting reaction.[28] In a recent study by Malacarne-Zanon et al.[36] it was reported that addition of 5 and 15% ethanol into model dental adhesives enhanced the degree of conversion; this phenomenon likely occurred by reducing the viscosity of resins to a level where molecular mobility increases with an increase in polymer chain segments.

Most of the recent studies involving the enhancement of hybrid layer integrity and longevity using GA derivatives were conducted by adding natural cross linkers directly into the adhesive bonding agents. Du et al.[37] reported that the incorporation of epigallocatechin-3-gallate (EGCG), which is a GA derivate into a dental adhesive, had favorable therapeutic effects such as increased antibacterial activity as well as improved microtensile bond strength. In another study by Neri et al.[38] the addition of EGCG into a self-etch bonding agent showed no adverse effects on the degree of conversion and flexure strength values. Hu et al.[39] investigated the physical properties of glass ionomer cements after the addition of EGCG and revealed improved flexural strength and surface hardness values in comparison with those of the control groups. However, in the present study, the mechanical properties of the tested RCs revealed unfavorable results associated with GA addition. This might be attributed to the inhibitory effect of phenolic compounds on free-radical polymerisation of methacrylate monomers.[40]

Conclusions

Addition of 0.5% and 1% GA did not affect setting time values in comparison with those of the control groups for both RCs. However, both flexural strength and hardness values of the tested resin cements significantly decreased with the addition of 0.5% and 1% GA. GA concentration and solvent type should be further optimised for a clinically successful modification.

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