ORIGINAL ARTICLE

Fluoride release/recharging ability and bond strength of glass ionomer cements to sound and caries-affected dentin

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Abstract

Objectives: The aim of this *in vitro* study was to evaluate the microtensile bond strength (μTBS) of different glass ionomer cements (GICs) on sound/caries-affected dentin and to assess the fluoride release/recharging ability.

Methods: After creating artificial caries lesions, teeth with sound and caries-affected dentin were assigned to be restored with one of the GICs: Ketac N100; GC Equia; GCP Glass Fill, and tested using a microtensile test. Ten of each material were prepared, and fluoride ion-release was measured at 10 different intervals. After 1.23% acidulated phosphate fluoride-gel immersion, fluoride re-release was then measured at the same intervals.

Results: In sound dentin, GC Equia exhibited significantly higher μTBS value ($P < 0.05$). The mean μTBS of Ketac N100 to caries-affected dentin was significantly lower than the other materials ($P < 0.05$). All materials showed maximal fluoride release at 24 h, GC Equia released the most fluoride.

Conclusion: The bond strength of GICs is affected by tooth substrate. High-viscosity GICs show higher initial fluoride release as well as greater fluoride recharge capacity.

Key words: Bond strength, caries-affected dentin, fluoride release, glass ionomer cements

Introduction

The current concept of restorative dentistry is characterized by the preservation of tooth structure during cavity preparation and less invasive approach for carious lesion treatment. Preserving the inner layer of intact, remineralizable, caries-affected dentin and removing only the outer layer of softened, caries-infected dentin is sufficient to ensure the caries-arrestment process, as long as a biological seal is created and maintained.[1] However, significant numbers of restorative failures, reported as recurrent caries, may result from residual caries.[2,3] Particularly in atraumatic restorative treatment, complete removal of decay is not always possible because hand instruments are used for excavation of the carious lesion or because incomplete caries excavation can occur in conventional cavity preparations, especially in children. Glass ionomer cements (GICs) have often been used as the restorative material of choice in these situations.[4]

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GIC was introduced in the 1970s by Wilson and Kent as a restorative material. These cements are the material of choice for the treatment of primary teeth for various reasons, including physical-chemical bonding to tooth substrate, satisfactory esthetic properties, biocompatibility, and continuous fluoride release to adjacent structures over a long period, inhibition of bacterial acid metabolism and activity, similar coefficients of thermal expansion of the tooth structure, and ease of clinical application.[5,6]

The chemical adhesions of GICs are achieved through ionic and polar interactions between hydroxyapatite and polycarboxylate radicals so that the latter displace the phosphate and calcium ions from the former.[7] This interaction is also considered to be beneficial in reducing hydrolytic degradation, thus enhancing restoration longevity.[8] However, bonding to caries-affected and sound dentin may vary due to differences in substrate, water content, permeability, and surface morphology.[9,10] Bond strength depends on the type of the material; however, there is limited information in the literature about the microtensile bond strength (μTBS) of GICs to both sound and caries-affected dentin in primary teeth.

Recurrent caries is the most common cause of failure of restorations, and several studies have demonstrated a lower incidence of recurrent caries is associated with the use of restorative materials that are capable of fluoride ion-release.[11,12] The fluoride release pattern of GICs is characterized by an initial rapid release, which may be related to high instability and erosion of GICs during the early setting period, followed by a rapid decrease in the rate of release.[13,14] The relatively low fluoride release after the initial reaction is thought to impair the ability of the materials to inhibit secondary caries because sustained, low levels of fluoride release may not achieve therapeutic levels. However, studies have also indicated that the GICs can be recharged with fluoride, which would provide a long-term caries inhibitory effect due to an increase in fluoride release.[15,16] These anticariogenic properties vary widely among different materials.

The type, concentration, and application frequency of the fluoride agent and the type and permeability of the restorative material are the main factors that affect the ability of the restoration to act as a fluoride reservoir.[17] Although several studies have been published regarding the fluoride release/recharging ability and μTBS of GICs,[5,6,9,13,17] there are no published reports that have compared the high-viscosity conventional GICs with nano-fluoride-/hydroxyapatite or resin-modified GICs (RMGICs) with nanoparticles. Therefore, the aims of this *in vitro* study was to assess μTBS of different types of GICs to sound and caries-affected dentin in primary teeth and the fluoride release/recharging ability of the tested materials. The null hypothesis was that there were no statistically significant differences for the evaluated parameters among the different materials tested.

Materials and Methods

In this study, three glass ionomer-based restorative materials (GC Equia, GCP Glass Fill, and Ketac N100) were used. The chemical composition, manufacturer, batch numbers, and application procedures of the materials are presented in Table 1. The present study was approved by the Medical Ethics Committee of the Izmir Katip Celebi University, under report number 2014/222.

The microtensile bond strength
Sample preparation
A total of 48 sound, caries-free primary second molars were selected for the study. Occlusal cavity preparations (7 mm × 5 mm × 2 mm deep) were performed using a fissure-type diamond bur (#856L; Brasseler USA Inc., Savannah, USA) that was operated at a high speed with oil-free, water-spray cooling by a single operator. The bur was replaced after every third preparation. The depth of the cavities was measured with a periodontal probe, and the width of the cavities was measured with a caliper. Following preparation, the specimens were divided into six groups according to the tested materials (three different GICs) and substrate (teeth with sound dentin and caries-affected dentin).

Creation of artificial caries
Half of the specimens ($n = 24$) were subjected to a cariogenic challenge to create artificial carious dentin lesions. The following protocol was used: First, the specimens were covered with two layers of an acid-resistant varnish (with the exception of inside the cavity preparations), and second, caries-affected dentin lesions were induced by a pH-cycling procedure. The demineralizing solution contained 2.2 mM $CaCl_2$, 2.2 mM NaH_2PO_4, and 50 mM acetic acid, adjusted to pH 4.8, and the remineralizing solution contained 1.5 mM $CaCl_2$, 0.9 mM NaH_2PO_4, and 0.15 M KCl, adjusted to pH 7. For 14 days, each specimen was cycled at room temperature for 8 h in 10 ml of demineralizing solution and then for 16 h in remineralizing solution. The solutions were renewed daily.

Restorative procedures
Teeth with sound and caries-affected dentin were randomly assigned to be restored with one of three materials: (1) RMGIC with nanoparticles (Ketac N100); (2) high-viscosity GIC (GC Equia); or (3) high-viscosity GIC with nanofluoro/hydroxyapatite (GCP Glass Fill). All materials were applied according to the manufacturer's instructions. For light-cured materials, a light emitting diode (LED) curing unit (Valo, Ultradent Products Inc., South Jordan, UT, USA) was used at an intensity of 1200 mW/cm² [Table 1]. Following the placement of the restorations and prior to obtaining the bond strength measurements, all specimens were stored in distilled water at 37°C for 24 h.

Specimen preparation

Teeth were sectioned in both mesial/distal and buccal/lingual directions across the adhesive interface using a low-speed diamond saw (Isomet, Buehler, Inc., Lake Bluff, IL, USA) to obtain specimens with a cross-sectional area of approximately 1.44 mm^2. The area of each specimen was measured with a digital caliper for the calculation of bond strength values. Then, each specimen was examined under ×40 magnification using a stereomicroscope to detect interfacial defects for exclusion. For each group, 30 bonded specimens were tested for the μTBS.

Microtensile bond strength test

The specimens were attached to a testing device (Shimadzu, Model AGS-X5 kN, Shimadzu Corporation, Kyoto, Japan) with cyanoacrylate glue and stressed at a crosshead speed of 1 mm/min until failure. The force in Newtons (N) required to displace the restoration was recorded and then the μTBS (in megapascal [MPa]) was calculated by dividing the imposed force (N) by the area of the adhesion (mm^2). The μTBS was calculated and expressed in MPa. Premature failures were included for statistical analysis as the mean of the lower value for each group and zero.

Assessment of failure modes

All debonded specimens were dyed with basic fuchsin and were observed with a scanning electron microscope (SEM) at ×200 magnification to identify the failure patterns. The failure modes were classified into three groups: (1) Adhesive failure between the restorative material and dentin, (2) cohesive failure within the dentin or in the restorative material, and (3) mixed fracture.

Statistical analysis

The μTBS data obtained from the six groups were analyzed using multivariate analysis of variance (ANOVA) and Tukey's *post hoc* test (α = 0.05). The failure mode was only qualitatively evaluated.

Fluoride release

Specimen preparation

A total of 30 disc-shaped specimens (10 of each material) were used in the study. The materials were prepared according to the manufacturer's instructions, and immediately after mixing, the materials were placed into a cylindrical Teflon mold 8 mm in diameter ×2.5 mm in height. Excess material was removed, and the specimens were then pressed between two Mylar-covered glass plates to obtain flat surfaces. The materials were polymerized according to the manufacturer's instructions with an LED light-curing unit. Following polymerization, each disc was removed from the mold, the dimensions of the specimens were measured with a digital caliper, and then were wet-ground with 600-, 800-, and 1000-grit silicon carbide abrasive paper for 10 s. Specimens were allowed

to set for an additional 24 h in a humid atmosphere at 37 ± 2°C.

Fluoride release analysis

Each specimen was immersed in a polyethylene tube containing 20 ml of deionized water (pH 6.5) (Milli Q plus, 18.2 M·cm, Millipore, New York, USA) and stored in an incubator at a constant temperature of 37°C. The fluoride ions released from each specimen were measured at 1, 2, 4, 8, 15, 22, 29, 36, 43, and 49 days. Following testing, the storage medium of each specimen was discarded, and specimens were transferred in a clean container, containing 20 ml of fresh storage medium. Concentrations of released fluoride ions were measured using a fluoride-specific ion electrode (WTW F800 Fluoride Combination Electrode, WTW Corporation, Weilheim, Germany) connected to a digital ion analyzer (WTW InoLab pH/Ion 7320, WTW Corporation, Weilheim, Germany). Prior to each measurement, the electrode was calibrated using standard solutions of 0.1, 1, 10, and 100 ppm fluoride. Recalibrations were performed every 10 measurements using the standard solutions. Measurements were performed by pipetting 3 ml of each sample solution into a clean plastic test tube, adding 3 ml of total ionic strength adjustment buffer II (TISAB II, WTW Corporation, Weilheim, Germany). Fluoride concentrations (ppm) of each solution were automatically displayed on the analyzer. After 49 days of ageing, the discs were removed from the polyethylene tubes and the specimens were individually immersed in 1.23% acidulated phosphate fluoride (APF)-gel (12,300 ppm fluoride, pH 3.2) (Sultan Topex APF, Sultan Dental Products, Englewood, NJ, USA) for 4 min in order to assess the fluoride recharge capability of each specimen. At the completion of fluoride immersion, each specimen was thoroughly rinsed with deionized water, dried with absorbent paper, and returned to its container, which was filled with 20 ml of fresh storage medium and stored at 4°C until analyzed. Fluoride re-release from each specimen was then measured at 10 analysis intervals on days 1, 3, 7, 14, 18, 22, 29, 36, 43, and 49 days following fluoride recharge, as described above.

Statistical analysis

A linear mixed-effects model was used, followed by pairwise comparisons of the repeated measurements and materials within each other considering Bonferroni adjusted nominal alpha levels as decision criteria. The major capabilities that differentiate a mixed model from a general linear model, such as repeated measures ANOVA, are that a mixed model handles correlated data and unequal variances.

In a mixed-effects model, random effects contribute only to the covariance structure of the data. The presence of random effects, however, often also introduces correlations between cases. Though the fixed effect is the primary interest in most studies or experiments, it is necessary to

Table 1: Chemical composition, application procedures and batch numbers of the tested materials

	Chemical composition	Application methods
Ketac N100 (#N493767, 3M ESPE Dental products, St. Paul, MN, USA)	Paste A: Fluoroaluminosilicate glass, silane-treated ZrO_2 silica, silane-treated silica, PEGDMA, HEMA, Bis-GMA, TEGDMA Paste B: Silane-treated ceramic, silane-treated silica, water, HEMA, acrylic/itaconic acid copolymer Fillers: 69% w/w (2/3 nano-fillers)	Apply Ketac N100 primer 15 s; gently air dry and light-cure the primer 10 s. Apply paste A and paste B on a mixing pad and mix for 20 s. Extrude the mixture in the cavity and light-cure 20 s
GC Equia (#1301251, GC Int., Tokyo, Japan)	Fluoroaluminosilicate glass, distilled water, polyacrylic acid	Etch for 20 s; gently air dry, shake the capsule, mix the capsule 10 s at high speed, remove from the mixer and make two clicks to prime the capsule then syringe. Extrude the mixture in the cavity. Dispense GC Fuji Coat LC and light-cure
GCP Glass Fill (#7211953, GCP Dental, Vianen, the Netherlands)	Nanoparticles glass, nanofluoro/hydroxyapatite, liquid silica	Clean the cavity with EDTA, gently air dry, shake the capsule, mix the capsule 10–15 s with high-frequency mixers and make two clicks to prime the capsule then syringe. Extrude the mixture in the cavity. Dispense GCP Gloss and light-cure
GCP Gloss (#1307076, GCP Dental, Vianen, the Netherlands)	Modified polysiloxanes	Dispense some drops of GCP Gloss onto a mixing pad and apply with a disposable brush, a thin layer of the coat to the fully built-up filling or sealant. Heat up each surface of the filling for 60-90 s
Fuji Coat LC (#1310241, GC Int., Tokyo, Japan)	Multifunctional urethane methacrylate, aliphatic dimethacrylate, methyl methacrylate, tertiary amine	Apply with a brush to all exposed glass ionomer surface and light-cure for 10 s
Gel Etchant 20% polyacrylic acid (#1305271, GC, Tokyo, Japan)	20% polyacrylic acid, distilled water, aluminum chloride hydrate	Etch for 10 s, rinse thoroughly with water and dry

EDTA=Ethylenediaminetetraacetic acid; UDMA=Urethane dimethacrylate; TEGDMA=Triethylene glycol dimethacrylate; HEMA=Hydroxyethyl methacrylate; Bis-GMA=Bisphenol A glycidyl methacrylate; PEGDMA=Polyethylene glycol-dimethacrylate

adjust for the covariance structure of the data. Therefore, a first auto-regressive covariance structure matrix AR was selected for the analysis in this study. A linear mixed-effects model was implemented to test the differences among 3 materials, 2 conditions, and 10 repeated release measurements, and the three aforementioned factors were taken as fixed factors, whereas the subjects were taken as a random factor.

Results

Results of μTBS test between study groups in the sound and caries-affected dentin are presented in Table 2. The μTBS of GC Equia and Ketac N100 was significantly affected by substrate, and the μTBS values of caries-affected dentin were significantly lower than those of sound dentin ($P < 0.05$). However, there were no significant differences in μTBS between sound and caries-affected dentin for GCP Glass Fill ($P > 0.05$). In sound dentin, GC Equia exhibited the highest μTBS value, which was significantly different from that of the Ketac N100 and GCP Glass Fill ($P < 0.05$), and there was no significant difference between Ketac N100 and GCP Glass Fill ($P > 0.05$). The mean μTBS of Ketac N100 to caries-affected dentin was significantly lower than that of GC Equia and GCP Glass Fill ($P < 0.05$), while the differences between these two groups were not significant ($P > 0.05$).

Data regarding the failure mode of debonded specimens are summarized in Table 3. Ketac N100 showed a higher

Table 2: Means of μTBS values and SD for each material and substrate

Substsrate	Mean (SD)		
	GC Equia	GCP Glass Fill	Ketac N100
Sound dentin	12.02 (2.72)[A,a]	7.56 (2.03)[A,b]	9.14 (3.71)[A,b]
Caries-affected dentin	8.58 (2.32)[B,a]	6.86 (1.65)[A,a]	3.50 (3.82)[B,b]

*Uppercase letters indicate statistically significance differences within columns ($P<0.05$); *Lowercase letters indicate statistically significance differences within rows ($P<0.05$). SDs=Standard deviations; μTBS=Microtensile bond strength

Table 3: The distribution of specimens evaluated in scanning electron microscope regarding failure modes

Group	Structure	Adhesive fracture	Cohesive fracture	Mix fracture
Ketac N100	CAD	22	4	4
	SD	15	9	6
GC Equia	CAD	12	12	6
	SD	8	12	10
GCP Glass Fill	CAD	11	14	5
	SD	9	15	6

CAD=Caries-affected dentin; SD=Sound dentin

number of specimens with adhesive failure compared with other materials, while GC Equia and GCP Glass Fill showed a higher number of specimens with a cohesive failure compared with the material. The predominant failure mode in sound and caries-affected dentin was an adhesive failure. Representative SEM images are showed in Figures 1-4.

Table 4: Fluoride release from glass ionomer materials (ppm) mean (SD)									
Groups	Day 1	Day 2	Day 4	Day 8	Day 15	Day 22	Day 29	Day 36	Day 43	Day 49
Ketac N100	1.650[A,a] (0.391)	0.642[A,b,c] (0.129)	0.724[A,b,c] (0.197)	0.750[A,b,c] (0.264)	0.876[A,b,c] (0.275)	0.696[A,b,c] (0.128)	0.709[A,b] (0.141)	0.557[A,b,c] (0.127)	0.587[A,c] (0.115)	0.641[A,b,c] (0.213)
GC Equia	30.576[B,a] (6.064)	18.148[B,b] (3.238)	20.191[B,b] (4.752)	19.114[B,b] (5.172)	17.631[B,b,c] (5.333)	12.783[B,c] (2.755)	10.788[B,d] (2.285)	7.605[B,e] (1.720)	7.380[B,e,f] (1.610)	6.589[B,f] (1.440)
GCP Glass Fill	15.974[C,a] (12.497)	10.407[C,b] (0.886)	11.525[C,b] (1.462)	11.746[C,b] (1.608)	10.768[C,b,c] (2.751)	7.681[C,c,d] (1.525)	6.099[C,d,e] (1.614)	5.319[C,e] (0.686)	5.199[C,e] (0.561)	4.744[C,e] (0.766)

*Capital letters indicate statistically significance differences within columns ($P<0.05$); *Lowercase letters label statistically significance differences within rows ($P<0.001$). SD=Standard deviation
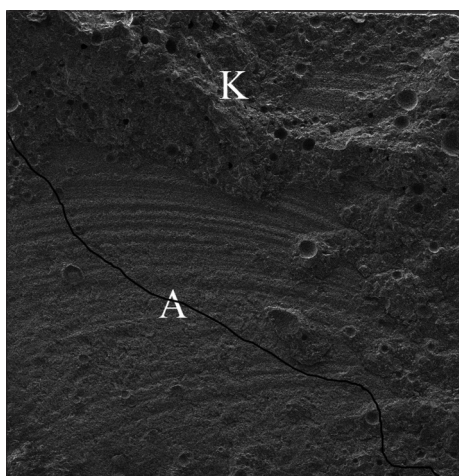

Figure 1: Mixed failure specimen in the GC Equia/Sound dentin group (×200). K = Cohesively failed region within GC Equia. A = Adhesively failed region
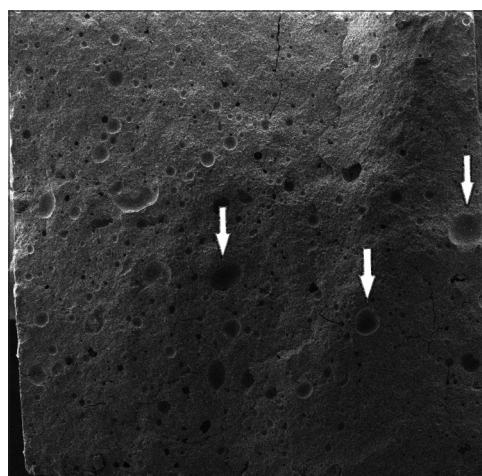

Figure 2: Cohesively failed specimen in the GCP Glass Fill/caries-affected dentin group (×200). The white arrows indicate air bubbles
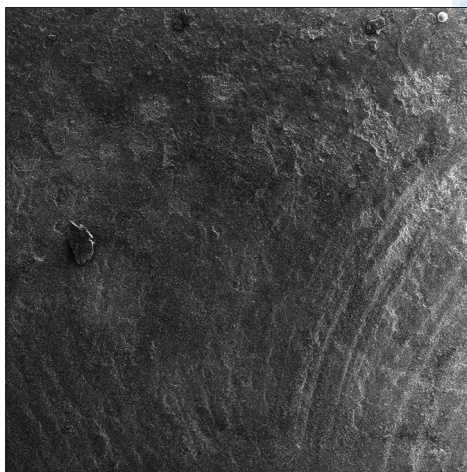

Figure 3: Scanning electron microscope image (×200) of a specimen indicates an adhesive failure in the Ketac N100/caries-affected dentin group
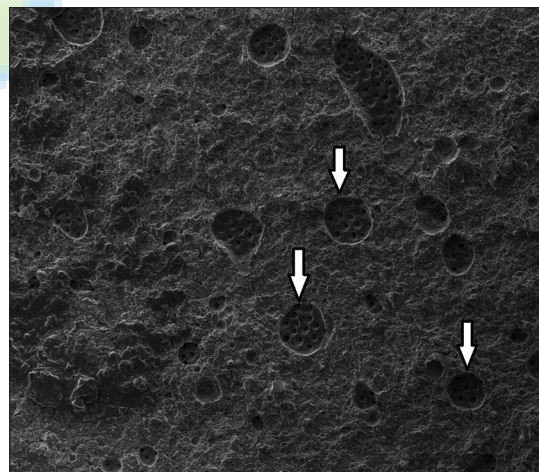

Figure 4: ×1000 image of a specimen in the mixed groups. The white arrows show exposed dentin surfaces

The mean ± standard deviation (SD) of fluoride release from each material before exposure to APF-gel are shown in Table 4. For all materials, the greatest amount of fluoride release occurred at 24 h. Fluoride release decreased with time, but continued throughout the entire 49-day test period up until recharge. On each day of testing, significant differences were observed between all of the materials in the following order: GC Equia > GCP Glass Fill > Ketac N100 ($P < 0.05$). The mean (±SD) amount of fluoride released from each material after exposure to APF-gel is shown in Table 5. For all materials, an increase in fluoride release occurred at 24 h following exposure to fluoride gel; however, after 24 h, the fluoride release levels of the materials decreased sharply. GC Equia released significantly more fluoride than the other materials during the first 7 days after fluoride gel exposure ($P < 0.05$). Ketac N100 released

Table 5: Fluoride rerelease from glass ionomer materials (ppm) mean (SD)

Groups	Day 1	Day 3	Day 7	Day 14	Day 18	Day 22	Day 29	Day 36	Day 43	Day 49
Ketac N100	1.066[A,a] (0.222)	0.254[A,b] (0.084)	0.463[A,c] (0.104)	0.561[A,c,d] (0.108)	0.500[A,c,d] (0.161)	0.491[A,c,d] (0.119)	0.617[A,d] (0.113)	0.609[A,d] (0.091)	0.506[A,c,d] (0.159)	0.448[A,c,d] (0.057)
GC Equia	7.643[B,a] (2.197)	3.131[B,b,d,e] (0.970)	4.946[B,c] (1.280)	4.491[B,b,c,d,e] (1.342)	3.268[B,d] (1.020)	4.061[B,b,c,d,e] (0.552)	4.181[B,c] (0.703)	3.622[B,b,c,d,e] (0.716)	3.647[B,b,c,d,e] (0.739)	3.405[B,d,e] (0.909)
GCP Glass Fill	4.543[C,a] (3.072)	1.944[C,b] (0.303)	3.356[C,a,d,e,h] (0.676)	3.731[B,a,d,e] (0.668)	2.288[C,b,c,f] (0.505)	2.706[C,b,c,f] (0.322)	3.311[C,c,g,h] (0.394)	3.172[B,d,e,g] (0.429)	2.992[C,e,f,g] (0.455)	3.030[B,e,f,g] (0.297)

*Capital letters indicate statistically significance differences within columns ($P<0.05$); *Lowercase letters label statistically significance differences within rows ($P<0.001$). SD=Standard deviation

significantly less fluoride than the other materials at all time points tested ($P < 0.05$).

Discussion

GICs are widely used in dentistry because they possess a variety of suitable properties.[5,6,18] Chemical diffusion-based adhesion to enamel and dentin tissues and fluoride release are some of their important properties. Over time, several modifications have been made to glass ionomer material formulations which have led to improved materials and, consequently, clinical benefits. The latest innovations are glass ionomer materials that are based on the nanotechnology or contain the addition of fluoroapatite/hydroxyapatite. Before these materials can be tested in clinical studies, it is essential to evaluate the performance of these newly produced glass ionomers in the laboratory. Therefore, the primary objective of this study was to evaluate µTBS and fluoride release/recharge capacity of three different content glass ionomer restorative materials *in vitro*. Based on the results of this study, the null hypothesis was rejected since differences in the µTBS and fluoride release/recharge capacity were observed among the different materials tested.

µTBS testing is an important method for assessing the benefits of restorative materials used for tooth restorations.[5,6,8,9] Since bonding to sound and caries-affected dentin may reveal differences in materials,[5,6,8,9] in this study, the µTBS of the three different GICs to sound and caries-affected dentin was investigated. A collection of caries-affected primary tooth dentin specimens is difficult because of the small dimensions and proximity of pulp. Therefore, to standardize across samples, artificial carious dentin was used, and artificial carious lesion induction in sound primary teeth was performed. Artificial carious lesions are advantageous because they create flat bonding surfaces, and the degree of demineralization of the dentin is standardized.[19] Moreover, the µTBS test on cross-sectional specimens was chosen in the current study to reduce specimen variability and to improve the validity of results. Because of the premature failure occurred when tried to obtain 1 mm × 1 mm dimension slabs, 1.2 mm × 1.2 mm dimension slabs were obtained in this study. The thicknesses of the slices were recorded using a digital caliper and the bond strength

expressed in MPa at failure was calculated by dividing the load in Newtons by the area of the bonded interface. Thereby the bonded interface differences were eliminated.

In the present study, significant differences were observed in the µTBS values in sound and caries-affected dentin for GC Equia and Ketac N100, suggesting that GIC bond strength is affected by these two different substrates when applied to primary tooth dentin. Although the mean µTBS values between the sound and caries-affected dentin of GCP Glass Fill is not significant, a numerically lower bond strength value was observed in caries-affected dentin. These findings are consistent with the findings of studies by Palma-Dibb *et al.*, Choi *et al.*[20,21] The chemical adhesions of GICs are achieved through ionic and polar interactions between hydroxyapatite and polycarboxylate radicals so that the latter displace the phosphate and calcium ions from the former.[7] Choi *et al.* [20] suggested that the lower µTBS of GICs to caries-affected dentin was caused by the chemical bonding mechanism between the calcium ion of hydroxyapatite and carboxyl groups of the polyalkenoic acid, that is, the loss of calcium ions through demineralization in the caries-affected dentin groups reduced the opportunity for bonding between calcium ions and the carboxyl groups.[21]

To improve the adhesion of RMGICs, some authors have suggested that the dentin surface should be treated with agents that promote smear layer removal or modification.[22-24] For example, Cardoso *et al.* reported that RMGIC bonds to dentin by micro-mechanical and chemical bonding mechanisms; however, micro-mechanical adhesion does not play an important role in the bonding mechanism of RMGIC and chemical interaction remains the primary bonding mechanism.[22] The RMGIC investigated in this study used a cured primer before the restoration, which may increase the bond strength to sound dentin. However, in caries-affected dentin, most of the dentin tubules are occluded by mineral deposits[25] and the presence of these mineral crystals may prevent resin tag formation during the bonding process. In support of this claim, in the caries-affected dentin that used Ketac N100, a total of 14 specimens failed during specimen preparation (during the sawing procedure and during cutting of the dentin base). The bond strength value of all specimens that failed prematurely was considered to be 0. The high number of

premature failures in the caries-affected dentin groups may reflect the weak bond strength of the material.

Based on the results of the present study, in the caries-affected dentin, GC Equia, and GCP Glass Fill showed similar and higher μTBS values compared with Ketac N100, respectively. In sound dentin, the GC Equia had the highest μTBS values when compared with the other glass ionomer-based materials. The different results between two conventional GICs may be explained by an inadequate micro-mechanical retention between the restoration and tooth structures, which was caused by a reduced etching capacity of surface treatment. Before the application of GC Equia, the dentin surface was treated with polyacrylic acid, and EDTA was used before the application of GCP Glass Fill. Studies indicated that, with high-viscosity GIC, the prior use of polyacrylic acid followed by washing can increase the bond efficiency.[26] This treatment creates a superficial cleaning and a partial demineralization, which enhances the likelihood of chemical and micro-mechanical interactions between the GIC and the hydroxyapatite. The superficial demineralization is important, especially for GIC, as its bond mechanism is also based on the mineral content of the tooth.[26]

Because of its better mechanical properties and the formation of a hybrid-like layer, most studies have shown that the mean bond strength of RMGICs to sound dentin is significantly higher than that of conventional GICs.[6,23,24,27] In this study, the mean bond strength of an RMGIC (Ketac N100) to sound dentin was significantly lower than that of GC Equia and similar to that of GCP Glass Fill. The nano-RMGIC Ketac N100 is based on nanotechnology and two-thirds of the filler is composed of nanofillers. The μTBS results obtained from Coutinho *et al.* showed that this nano-RMGIC bonded as effectively to dentin as a high-viscosity GIC, but less effectively than the other RMGICs. In a transmission electron microscopy analysis of nano-RMGICs, Coutinho *et al.*[26] reported the presence of a tight interface between the enamel and dentin without surface demineralization and hybrid-layer formation. Thus, the bonding mechanism of nano-RMGICs should be attributed to micro-mechanical interlocking provided by the surface roughness, most likely combined with a chemical interaction mediated by its acrylic/itaconic acid copolymers.[27]

Failure mode classifications of the debonded specimens are an important part of bond strength testing. This process includes an analysis of the correlation between bond strength and failure mode. In this study, adhesive failure samples were observed in the caries-affected dentin more frequently than sound dentin that finding is consistent with the statistical results. The cohesive failure patterns were observed predominantly in the conventional GICs. The predominance of a cohesive failure mode of glass ionomer materials has often been interpreted as an indication that the bond strength values represent a lower tensile strength of the tested GIC material rather than its true adhesive bond strength to dentin.

As the fluoride release from dental materials is directly related to their cariostatic effect, several studies have been performed on fluoride release and recharge capacity of the materials tested herein.[11,13-17] Fluoride release from a GIC is a complex process and affected by intrinsic and extrinsic factors. Intrinsic factors include formulation, powder/liquid ratio, specimen geometry, temperature, mixing time, solubility or porosity of the material, surface treatment, and finishing.[13-17] Extrinsic factors include type and pH of storage medium, experimental design, the environmental temperature, and analytical methods. In this study, specimen geometry, surface treatment, environmental temperature, the type and pH of the storage medium, the experimental design, analytic method, and finishing were standardized for all materials.[13-17,28] Deionized water, saliva or pH-cycling models are preferred mediums to evaluate the fluoride release from dental materials.[17,28] Although saliva or pH-cycling models could better simulate the oral environment, deionized water is a medium that reflects well the fluoride release of the materials without the confounding influence of minerals or organic molecules which might be presented in saliva or pH-cycling solutions. Dionysopoulos *et al.* reported that the fluoride release pattern from restorative materials were similar in solutions simulating pH-cycling and deionized water.[28]

In the present study, all the tested materials released measurable quantities of fluoride during the 49-day period prior to APF solution treatment. However, there were large variations in the amount of fluoride released from the tested materials. Fluoride release from GICs occurs by means of three mechanisms: Surface loss, diffusion through pores and cracks, and bulk diffusion.[13,14] All fluoride-containing dental materials investigated in this study released the greatest amount of fluoride ions on the 1st-day. Fluoride continued to be released in relatively low amounts from day 2 until day 49. The high level of fluoride release from GIC materials on day 1 was probably due to an initial "burst" of fluoride release from the glass particles. The burst release is attributed to the reaction of the polyalkenoic acid with the fluoride-containing glass particles during the setting reaction and also to the rapid dissolution of fluoride from the outer surface into the solution.[13,14] The slower release of fluoride during subsequent days has been attributed to the slower dissolution of glass particles through cement pores and fractures. Bulk fluoride diffusion occurs during the maturation period as a consequence of contact between the GIC material with the storage medium.[13]

Among the restorative materials, GC Equia released significantly more fluoride than the GCP Glass Fill and Ketac N100. In general, a direct relationship exists between the amount of fluoride present in the cement and the amount of fluoride released. In comparison to GC Equia and GCP Glass Fill, which are conventional GICs, Ketac N100 released less fluoride. To date, several studies have compared the fluoride release pattern of RMGICs and conventional high-viscosity GICs. The fluoride release potential of RMGICs is equal to that of conventional GICs.[13-17] However, several variables affect the fluoride release capability of RMGICs. These include the type and amount of resin used for photochemical polymerization reaction and the presence of fluoride compounds and their interaction with polyalkenoic acids.[14,15,28] Our findings are consistent with the results of Dionysopoulos *et al.*, which indicated that Ketac N100 released less fluoride when compared with a conventional GIC.[28]

Inhibition of secondary caries is usually associated with a continuous fluoride release from the restorative materials.[11,12] Reductions in the rate of fluoride release and the recharging capacity of the fluoride-releasing materials become interesting and important issues, especially for patients with a high risk of caries. The fluoride re-release patterns of GICs tested in the present study agreed with the results of other studies: All the tested materials were found to recharge with fluoride and re-release again.[13-17] After fluoride recharge, fluoride release increased in the first 24 h, but after this period, the amount of fluoride release from all materials experienced a sharp drop. However, fluoride recharging from APF solution caused conventional GICs to exhibit significantly higher fluoride re-release than RMGICs. The precise mechanism of fluoride uptake by the restorative materials is not fully understood. The diffusion of fluoride into the material, material composition, and differences in surface energy may affect fluoride recharge and release ability.[11,13,14] It has been suggested that the recharging ability of GICs is dependent on the glass component, particularly the structure of the hydrogel layer around glass filler particles.[11-16] Additionally, GICs are significantly more porous and permeable than RMGICs, thus enhancing fluoride release. These findings indicated that materials with higher initial fluoride release have higher recharge ability, which are consistent the results of other studies.[15,18,28]

Conclusion

1. The sound and caries-affected dentin substrates alter the μTBS of GIC materials.
2. Conventional GICs have higher fluoride release and recharge ability than RMGICs.

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