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An in vitro study on the maturation of conventional glass ionomer cements and their interface to dentin

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ABSTRACT

The objective of the study was to investigate the influence of long-term storage (up to 1 year) and coating on the variation of micro-mechanical properties of four conventional restorative glass ionomer cements (GICs) within 3.5 mm deep class I cavities. Four commercially available GICs (Riva Self Cure (SDI), Chem-Fil Rock (Dentsply), Fuji IX Fast and Fuji IX GP Extra/Equia (GC)) were applied to 100 teeth. In each tooth, two similar 3.5 mm deep class I cavities were prepared and filled with the GICs, with and without resin coating. The samples were stored in artificial saliva at 37 °C for 1 week, 1 month, 3 months, 6 months and 1 year. The variation in mechanical properties (indentation modulus (*E*) and Vickers hardness (*HV*)) were determined in 100 μm steps starting from the filling surface, through the intermediate layer in between dentine and GIC, and ending 100 μm in dentin. *HV* and *E* were strongly influenced by the material ($P < 0.05$, partial eta-squared $\eta_P^2 = 0.31$ and 0.23) but less by aging duration ($P < 0.05$, $\eta_P^2 = 0.02$ and 0.12) and resin coating ($P < 0.05$, $\eta_P^2 = 0.02$ and 0.03). The depth of measurement (0–2 mm) has no influence on *HV* ($P = 0.789$). *HV* shows a gentle increase over the 1 year storage period ($P = 0.002$). A ~300 μm GIC zone at the areas close to dentin with weaker properties as those measured in dentin or GIC was identified in all fillings, irrespective of the presence of coating, and at all storage periods. The thickness of this zone is more strongly influenced by storage ($P < 0.05$, $\eta_P^2 = 0.081$) than by material type ($P < 0.05$, $\eta_P^2 = 0.056$), while coating showed no influence ($P = 0.869$). Filler morphology and dimension were similar to upper parts of the GIC filling; however, the amount of low cations was higher. We concluded that the development of an intermediate layer in between dentine and GIC with lower mechanical properties might be responsible for the bond quality of GIC to dentine. Moreover, class I GIC restorations are unlikely to feature constant mechanical properties throughout the cavity, regardless of conditions such as aging and coating.

1. Introduction

Glass ionomer cements (GICs) are used in modern dentistry as temporary restoration materials for cavities away from high-stress locations. Due to their ability to adhere to moist enamel and dentin without necessitating an intermediate agent and their anti-cariogenic properties such as long-term fluoride release, as well as good biocompatibility and low coefficient of thermal expansion, they have proved to be a reliable filling material [1–5].

In order to improve mechanical properties to achieve acceptable longevity in high-stress regions, manufacturers supply resin coatings and adjust the composition and shape of the glass fillers, as well as the composition and combination of the polyacrylate acids. It has already been shown that a resin coating is able to produce an improved performance in GICs [6,7]. The balance between water uptake and water loss represents a decisive factor for better clinical results of GIC fillings, which might be achieved by an immediate covering of the immature GIC surface, thus limiting water movement across the surface [6]. In former studies it was found that resin coating is a protective component which should be applied on GIC restorations [5,8]. However, micro-mechanical properties such as Vickers hardness (*HV*) and indentation modulus (*E*) obviously do not profit from surface protection, as found in a recent study [8]. Micro-mechanical properties are thus presumably not affected by aqueous solutions with a neutral pH as artificial saliva and distilled water. However, mechanical strengths change during long-term staging [9–11]. A previous study [12] using aluminum magic angle spinning-nuclear magnetic resonance (Al MAS-NMR) spectroscopy over a 1 year period concluded that the composition of the original glass has a considerable effect on the cement's setting reaction. There the formation of six-coordinate aluminum (Al) was also observed; this cross-links the carboxyl groups in the polyacrylic acids. The six-coordinate Al results from a conversion of four coordinate-Al, leading to an increasing proportion of six-coordinate Al with setting time, and thus to the

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increasing formation of octahedral Al ions, which cross-link the carboxyl groups of polyacrylic acids. In addition, this study showed that phosphorus has a strong influence on the setting reaction of the cement [12].

A few studies have described mechanical properties over several time periods or in varying storage solutions [13–16,9], but not necessarily at varying locations of an in vitro sample, thus neglecting the influence of dentin and enamel. In particular, the interface between GIC and dentin might reveal a specific mechanical behavior due to chemical interactions between both zones. Investigations on the interfacial occurrences between GIC and dentin, such as ion-exchange processes [17,18], have been carried out between dental cements and hydroxyapatite (dentin and enamel) as those exchanges may influence the bond strength between cement and dentin. In GIC fillings, it was found [19] that a dissociation of cross-linking of polycarboxylate chains might occur due to diffusion of hydrogen ions. Those ions are likely to exchange with matrix-forming cations, inducing dissociation [19]. The diffusion process might be controlled by the difference in the concentration of ions in the proximate surroundings and the GIC matrix [19]. X-ray photoelectron spectroscopy of the GIC–dentin interface made it possible to reveal atomic ratios between the elements found in the interface and those found in the GIC and dentin. A varying atomic ratio between GIC and the interfaces indicates ion diffusion. Ca/Si and Ca/C ratios show that the dentin interface consists of elements of dentin and glass ionomer cement, while the GIC interface is mainly built up of elements of the GIC and a small amount of calcium, but no peptidic nitrogen. This XPS study showed that an interphase is formed by reciprocal diffusion of the different elements comprising the GIC and the dentin, but without collagen [20]. Thus, one should ask if the mechanical properties in these interfacial areas might alter significantly during long-term storage due to change within the slow diffusion of elements of the GIC through the dentin during long-term interactions. The aim of our study was therefore to pursue changes in the mechanical properties of four different GICs and their interactions with dentin, as a function of coating application and storage duration.

The null hypotheses tested were: (i) aging for 1 year and resin coating would not influence the *HV* and *E* of the GIC or the GIC–dentine interfacial areas; (ii) within one material, different depths of the cavity would reveal similar results for *HV* and *E*; (iii) *HV* and *E* would not be influenced by the different compositions of the GICs.

2. Materials and methods

Four conventional restorative GICs – Riva Self Cure, Fuji IX GP Fast and Fuji IX GP Extra (Equia) and ChemFil Rock – were selected (Table 1). ChemFil Rock is a new conventional GIC incorporating zinc in the chemical formulation of filler. The GICs were all encapsulated.

A corresponding light-cured resin coating for each material was chosen. There is no indication given by the manufacturer of ChemFil Rock concerning the application of resin coating. Nevertheless, in order to confront every material with the same conditions an experimental resin coating supplied by the same manufacturer was applied also on this material (Table 1).

100 extracted non-carious molars were collected and stored in a sodium azide solution (3%). Prior to preparation the teeth were thoroughly cleaned with distilled water. Two standardized cone cavities 3.5 mm in depth and 3 mm in diameter at ground level were prepared in each tooth by using a diamond bur with the shape of a truncated cone of 3 mm diameter at its bottom (Fig. 1).

The encapsulated GICs were mixed by rotating in a RotoMix (3 M-ESPE, Seefeld, Germany) apparatus and the GIC fillings were prepared at room temperature according to the manufacturer's instructions. The cavities were cleaned with water and dried gently before the GIC was applied. One of the two cavities received an additional resin coating, light-cured for 20 s with an irradiance of 1100 mW cm^{-2} (Mini LED, SATELEC SED-R, Merignac, France). A total of 25 teeth for each material were prepared, with five teeth for one aging period. The samples were than stored in artificial saliva (pH 6.9) with the composition of 1.2 g KCl, 0.84 g NaCl, 0.26 g K_2HPO_4 and 0.14 g $CaCl_2{\cdot}2H_2O$ per 1000 ml distilled water at 37 °C for 1 week, 1 month, 3 months, 6 months and 1 year. The artificial saliva was changed every day for samples stored for 1 week and 1 month, while for the long-term storage specimen, weekly renewals took place.

Before measuring, the teeth were cut mesio-distally, through the center point of the cavities by a circular saw (Isomet Low Speed Saw, Buehler, Lake Bluff, USA), to produce a cross-sectional area. The slices were ~2 mm thick. The surface of the cross-section was wet-ground with 2500 and 4000 grit silicon carbide paper (FEPA, Hermes, Hamburg, Germany). The samples were than fixed on an object slide and the mechanical properties *HV* and *E* were determined by means of an automatic micro-hardness indenter (Fischersope H100C, Fischer, Germany). The measurements were done in 100 μm steps starting from the surface of the filling in the middle of the filling and ending ~100 μm within the dentin. For each cavity, coated and uncoated, two operated sequences of indentation were performed. The integrated light microscope was used to take images of the cavity bottom and margin with 40× magnification. For statistical analysis the values measured at different positions, namely surface (position 0), at 1 mm depth (position 1), at 2 mm depth (position 2), as well as the intermediate layer in between dentine and GIC (position 3) and dentine (position 4) were considered.

The measurements were carried out under force control: the test load increased and decreased at a constant speed between 0.4 and 500 mN. The load and the penetration depth of the

Table 1
Materials, manufacturer and chemical composition of glass ionomer cements (all encapsulated) and coating materials.

Glass Ionomer Cement	Manufacturer	Composition
Riva Self Cure; Lot: B1004281	SDI Limited, Victoria, AUS	Fluoro- aluminosilicate glass Polyacrylic acid + Tartaric acid, Polyacrylic acid
ChemFil Rock; Lot: 1005004004	Dentsply, Konstanz, GER	Calcium-aluminum-zinc-fluoro-phosphor-silicate glass, Polycarboxylic acid, Iron oxide pigments, Titanium dioxide pigments, Tartaric acid, Water
GC Fuji IX GP Fast; Lot: 1005211	GC Europe N.V., Leuven, BEL	Alumino-fluoro-silicate glass, Polyacrylic acid, Distilled water, Polybasic carboxylic acid
GC Fuji IX GP Extra (Equia); Lot: 1005281	GC Europe N.V., Leuven, BEL	Polyacrylic acid, Alumino-silicate glass, Distilled water
Coating		
Riva Coat; Lot: 091103	SDI Limited, Victoria, AUS	Acrylic monomer
Seal&Protect TF; Lot: MTO-3-27-1	Dentsply, Konstanz, GER	Di- and trimethacrylate, Acetone, Dipentaerythritol penta acrylate monophosphate
GC Fuji Coat LC; Lot: 1005061	GC Europe N.V. Leuven, BEL	Methylmethacrylate, Multifunctional methacrylate, Camphorquinone
GC G-Coat Plus; Lot:1004091	GC Europe N.V. Leuven, BEL	Methyl methacrylate, Colloidal Silica, Camphorquinone

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