Shear bond strength of bio-active cement versus self-adhesive resin cement with enamel and dentin when bonded to zirconia in wet and dry conditions (in-vitro study)

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Abstract: ACTIVA bio-active cement is resin-modified glass-ionomer cement that was introduced for re-mineralizing of the disintegrated joint. Due to the lack of data about this bioactive cement, this study evaluated the shear bond strength of bioactive cement and self-adhesive resin cements to tooth substrate (enamel and dentin), in wet and dry conditions when bonded to zirconia. In this study, 40 extracted mandibular molars caries-free were collected. Each tooth was cut by diamond disk (bucco-lingual section) to expose the dentin and enamel. Teeth were randomly divided into two groups enamel and dentin (n=20). Each group was further divided into two subgroups; wet and dry samples (n=10). Each subgroup was divided according to the cement used, into resin cement or bioactive cement (n=5) and bonded to zirconia. Shear bond strength was determined, after thermo-cycling. Statistical analysis of the results showed that there was no statistically significant difference between shear bond strength of the two cement types either with wet enamel or dentin. With both cements, enamel showed statistically significantly higher mean shear bond strength than dentin. It was concluded that, both cements showed comparable results with significant superiority to the self-adhesive resin cement.

1. Introduction

Dissolution of cement is a risk factor that diminishes marginal integrity of the cast restorations and results in bacterial ingress and secondary decay which leads consequently to its failure. Therefore, bioactive cement was introduced to overcome the shortcomings of the contemporary cements to increase the success rate of restorations [1]. The principle of bioactive restorative materials is not recent it existed since the introduction of glass-ionomer into the market that had the ability to release fluoride and resist secondary decay but lacked strength, toughness and instability due to water sorption [2]. Then resin modified glass ionomer (RMGI) that combines the advantages of glass ionomer cement and resin cement, was developed but had questionable biocompatibility and limited bioactivity [3].

ACTIVA bio-active cement was introduced that comprises of RMGI besides modified resin matrix (Blend of diurethane, other methacrylate with modified polyacrylic acid and bioactive nanoparticles),
that is capable of penetrating dentin through micro-mechanical interlocking. It forms a combined ionic bond between polyacrylic acid and hydroxyapatite and a micro-mechanical interlocking with dentin tubules and collagen. It does not elicit post-operative sensitivity and can tolerate PH cycles, leach and recharge calcium, phosphate and fluoride according to the manufacturer \(^4\). In 2015, Pameijer et al. \([6]\) affirmed that Activa bioactive RMGI flexural strength is equivalent to that of flowable composite and greater than that of glass ionomer cement (GIC), concerning biocompatibility Zemener et al. \([6]\) conducted test to assess sealing capability of the new RMGIC and compared it to other four luting agents for complete cast crown cementation and concluded that Activa demonstrated the lowest micro leakage scores. In 2017, Korkut et al. \([7]\) found that ACTIVA cement showed better mechanical and physical properties that was attributed to its content of the bioactive nanoparticles because it fills the matrix gap and the cracks formed under stress are smaller and fewer.

RelyX unicem cement is self-adhesive resin cement with a multifunctional phosphoric acid that required no pretreatment of the tooth or the restoration. This cement is moisture tolerant and capable of releasing fluoride the same as glass ionomer cements. RelyX Unicem cement is recommended for full-coverage restoration, including gold, porcelain-fused-to-metal, or all-ceramic/composite crown and untreated zirconia \([8]\).

Success of adhesive bonding in dentistry is multifactorial; it depends on type of substrate, type of adhesive substance, operator skills to bond the restoration successfully and humidity of the working area. Regarding the different substrates for instance, enamel is highly mineralized, which facilitates bonding procedure. On the other hand, dentin is not as of harmonious structure as enamel, it is composed of inorganic, organic components and water that demands ideal moisture conditions to ensure proper bonding by avoidance of collapse of the organic part of the dentin(collagen fibres) \([9]\).

Zirconia is a polycrystalline material with better mechanical and physical properties that is used in different dental prosthetisuch as, single crowns, adhesive bridges and fixed partial dentures. Bonding zirconia-based restorations is a critical issue; it cannot be etched with hydrofluoric acid because of its polycrystalline composition. It can be sandblasted which increases the mechanical bond strength. Another method of enhancing bond strength is, silane coupling agents with tribo-chemical treatment. Self-adhesive resin cements are used with zirconia restorations without a preceding step to prepare the surface \([10]\).

In general, it is difficult to control moisture in the oral cavity due to the presence of diverse contaminants like saliva, gingival fluid, blood and hand piece lubricant especially in sites near gingival margin \([11]\). This vitro study evaluated and compared the shear bond strength of the bioactive cement (Activa) with self-adhesive resin cement (RelyX) when bonded to enamel and dentin in dry and wet conditions and determined the mode of failure of each sample. The null hypothesis was that there is no difference between shear bond strength of the two cements with tooth structure, in dry and wet conditions and with enamel and dentin.

### 2. Materials and Methods

The tested cements Activa bioactive cement, Relyx self-adhesive resin cement and zirconia Aidite are shown in table 1.

### Table 1. The materials used in this study are listed.
### Material | Composition and description | Manufacturer | Batch number
--- | --- | --- | ---
Activa bio-active cement | Blend of diurethane and other methacrylates with modified polyacrylic acid (53.2%) Amorphous silica (3.0%) Sodium fluoride (0.90%) | PULPDENT, USA | 161007
RelyX Unicem Dual Cure Self-adhesive Resin cement. | In a capsule form it consists of: Powder: Glass Fillers, Silica, Calcium Hydroxide, Self-Curing Initiators, Pigments, Light-Curing Initiators. Liquid: Methacrylate Phosphoric Esters, Dimethacrylates, Stabilizers, Self-Curing Initiators, Light-Curing Initiators. | 3M ESPE, USA | 669862
Zirconia based ceramic block | ZrO$_2$, Y$_2$O$_3$, HfO$_2$, Al$_2$O$_3$, Na$_2$O Yttria-stabilized tetragonal Zirconia polycrystal blocks Shade A2 | Aidite (Qinhuangdao) Technology | S160716ATA2M-7 3D

#### 2.1. Tooth preparation
In this study, 40 caries free, non-identified mandibular molars were collected and stored in distilled water with 0.5% thymol for its antibacterial properties [12]. Teeth were sectioned from roots using diamond disc at the cement-enamel junction and were placed in a circular mold (polypropylene plastic tube) of 24 mm diameter and 1.5 cm height filled with epoxy resin (Kemapoxy, JM, Egypt). Epoxy resin was prepared by mixing p/m (3.5/1) by volume according to manufacturer instructions. They were randomly divided into two main groups enamel and dentin (n=20). For Enamel group buccal surfaces of the teeth were prepared by just scratching using a low speed diamond saw (Isomet 4000, Linear Precision Saw). For Dentin group, buccal surfaces of the teeth were prepared using the same diamond saw under copious water to flatten the buccal surface and expose 2 mm of the dentin surface [13]. Each group was divided into two subgroups wet and dry samples (n=10). Each subgroup was farther divided according to the cement used into self-adhesive resin cement as control group and bioactive cement (n=5) (figure 1).

![Figure 1. Samples grouping](image)

#### 2.2. Zirconia disc preparation
Zirconia rods were prepared from zirconia blocks (Aidite, Qinhuangdao Technology Co., LTD.), using computer numerical cutting machine (CNC), then were cut using a water-cooled diamond saw (Isomet 4000, Linear Precision Saw) at low speed. The amount of post-sintering shrinkage was considered, by preparing slightly oversized discs to reach their final dimension (3 mm diameter x 2.5 mm thickness) after sintering at 1450 °C and cleaned from debris (14). Zirconia discs were airborne-particle abraded using 50 μm Al₂O₃ at 2.5 bars of pressure (15 seconds) at adjusted distance of 10 mm before bonding.

2.3. Samples bonding
For subgroup samples planned to have a dry condition, they were dried with oil-free air for 10 seconds. For those to have a wet condition, water was applied once using a micro-brush disposable applicator (Microbrush International, Grafton, USA) for enamel and dentin samples. Excess water was removed with passive application of absorbent paper (Kleenex) only once (10). Samples of both groups (enamel and dentin) were bonded with bio-active cement or resin cement according to manufacturer instructions where the cement was injected on the previously prepared buccal surfaces of teeth. Then, zirconia discs were placed over the substrate by slight finger pressure, followed by a load of 5 kg applied using a loading device that was held for 10-15 seconds before light curing using (Optilux501, SDS Kerr, Danbury, USA), which was operated in standard mode (10). During load application, tack curing was applied and then excess cement was removed by a probe. Complete curing was proceeded for 20 seconds around the margins according to the manufacturer instructions.

2.4. Thermo-cycling and SBS testing
Samples were thermo-cycled for 3,000 thermal cycles between 5°C and 55°C temperature with 30 seconds dwell time at each temperature in distilled water in thermo-cycling unit which is equivalent to 3 months of clinical life (14). Shear bonding testing of all groups was measured using universal testing machine data recorded using computer software Bluehill version 3.3 at a cross-head speed of 0.5mm/min and load cell of 5000 Newton in a compression mode using a mono-bevel chisel of 0.5 mm thickness parallel to enamel and dentin surfaces as the shearing element (figure 2) [15]. Measurement of shear bond strength values were calculated by converting the loads into Megapascal (MPa) by dividing the maximum failure load Newton (N) by the bonding area (mm²) (16).

Figure 2. Shear bond testing using mono-bevel chisel.
Eight representative specimens from each sub-group were mounted on aluminum stubs with carbon glue after the critical-point drying process. The specimens were gold sputter-coated and analyzed using field-emission scanning electron microscopy (FE-SEM S-4100; Hitachi, Wokingham, UK) at 10 kV and a working distance of 15 mm and using magnification of 800 x and 1000 x.

2.6. Statistical analysis
Numerical data were explored for normality by checking the distribution of data and using tests of normality (Kolmogorov-Smirnov and Shapiro-Wilk tests). Data showed non-normal (non-parametric) distribution. Data were presented as mean, standard deviation (SD), median and range values. Mann-Whitney U test was used to compare between wet and dry conditions, the two cement types as well as the two substrates. The significance level was set at P ≤ 0.05. Statistical analysis was performed with IBM® SPSS® Statistics Version 20 for Windows.

3. Results

3.1. Shear bond strength
The mean, standard deviation and median for shear bond strength of the two cement types when bonded to enamel or dentin in two humid conditions were calculated. Comparison between shear bond strength values (MPa) of the two cement types is shown in table 2. Regarding wet condition; either with enamel or dentin, there was no statistically significant difference between shear bond strength of the two cement types. Regarding dry condition; there was no statistically significant difference in shear bond strength of the two cement types with dry enamel. Meanwhile, Rely X cement showed statistically significantly higher mean shear bond strength than Activa cement when bonded to dry dentin.

Table 2. The mean, standard deviation (SD) values and results of Mann-Whitney U test for comparison between shear bond strength values (MPa) of the two cement types.

| Humidity | Substrate | Rely X | Activa | P-value |
|----------|-----------|--------|--------|---------|
|          | Mean      | SD     | Mean   | SD      |
| Wet      | Enamel    | 6.15   | 2.27   | 3.89    | 1.44    | 0.095  |
|          | Dentin    | 1.26   | 0.66   | 0.63    | 0.35    | 0.421  |
| Dry      | Enamel    | 3.37   | 0.83   | 2.53    | 1.08    | 0.421  |
|          | Dentin    | 1.64   | 0.93   | 0.51    | 0.06    | 0.008* |

* Significant at P ≤ 0.05

Comparison between shear bond strength values (MPa) in wet and dry conditions is shown in table 3. Using Rely X cement, wet enamel showed statistically significantly higher mean shear bond strength than dry enamel. While there was no statistically significant difference between shear bond strength of wet and dry dentin. Using Activa cement, there was no statistically significant difference between wet and dry enamel or dentin. Either under wet or dry conditions and using both types of cements, enamel showed statistically significantly higher mean shear bond strength than dentin (table 4).

Table 3. The mean, standard deviation (SD) values and results of Mann-Whitney U test for comparison between shear bond strength under different humidity conditions.
Cement type    | Substrate | Wet | Dry |
|---------------|-----------|-----|-----|
|               |           | Mean| SD  | Mean| SD  | P-value |
| Rely X        | Enamel    | 6.15| 2.27| 3.37| 0.83| 0.032*  |
|               | Dentin    | 1.26| 0.66| 1.64| 0.93| 0.548   |
| Activa        | Enamel    | 3.89| 1.44| 2.53| 1.08| 0.310   |
|               | Dentin    | 0.63| 0.35| 0.51| 0.06| 0.151   |

* Significant at $P \leq 0.05$.

**Table 4.** The mean, standard deviation (SD) values and results of Mann-Whitney U test for comparison between shear bond strength of the two substrates.

Humidity | Cement type | Enamel | Dentin | P-value |
|----------|-------------|--------|--------|---------|
| Wet      | Rely X      | 6.15   | 1.26   | 0.008*  |
|          | Activa      | 3.89   | 0.63   | 0.008*  |
| Dry      | Rely X      | 3.37   | 1.64   | 0.032*  |
|          | Activa      | 2.53   | 0.51   | 0.008*  |

* Significant at $P \leq 0.05$.

**Figure 3.** SEM photomicrograph (magnification 1000x): Mixed failure in dry enamel surface with Activa cement

3.2. **Scanning electron microscopy**
ACTIVA bioactive cement samples demonstrated mixed failure when bonded to enamel. SEM photomicrograph showed clusters of cement debris failed to completely infiltrate through the enamel layer (figure 3). Meanwhile, samples showed adhesive failure when bonded to wet dentin surface with partial obliteration of the dentinal tubules with cement remnants (figure 4). ACTIVA bioactive cement samples showed mixed failure when bonded to dry dentin surface. SEM photomicrograph revealed partial obliteration of the dentinal tubules with cement remnants (figure 5).
Figure 4. SEM photomicrograph (magnification 800x): Adhesive failure in wet dentin surface with Activa cement.

Figure 5. SEM photomicrographs magnification (1000x): Mixed failure in dry dentin surface with Activa cement.

RelyX Unicem demonstrated mixed mode of failure when bonded to dry enamel. SEM photomicrograph revealed clusters of resin remnants on the enamel surface (figure 6). RelyX Unicem demonstrated adhesive failure when bonded to dry dentin. SEM photomicrograph showed partial obliteration of the dentinal tubules with no cement remnants (figure 7).
Figure 6. SEM photomicrograph (magnification 1000x): Mixed failure of RelyX Unicem in dry enamel.

Figure 7. SEM photomicrograph (magnification of 800x): Adhesive failure of Relyx Unicem in dry dentin.

4. Discussion
Dental cements are one of the factors that determine the success of indirect restorative restorations as they provide the required retention to dental surfaces and seal tooth restoration margins. Also incorporation of Nano-sized particles could decrease the setting time and enhance compression strength and elastic modulus [17]. Bond strength values obtained in this study were comparable to those found in another study; Hattar et al in 2015, that recorded nearly similar shear bond strength values with RelyX Unicem (6.81 MPa for enamel) [18]. This could be attributed to similar factors such as, bonding tooth substrate and cement type [19].

Regarding the effect of cement type: with dry dentin, RelyX cement showed statistically significant higher mean shear bond strength than Activa cement. Gerth et al. [20], in their study showed an increased chemical interaction of RelyX with Calcium from hydroxyapatite, which may explain the higher bond strength of this cement in this study.

Concerning the effect of wet and dry conditions, using Rely X cement with wet enamel showed statistically significantly higher mean shear bond strength than dry enamel which is in accordance with results obtained from Mushashe et al. [21]. It was pointed out that self-adhesive cement needs ionizing
medium for the chemical reaction to start and enhances the adhesive ability to interact with both substrates enamel and dentin. Similarly, Fursue et al. [22] found that all in one adhesive was affected positively by the presence of water and that was rationalized by the fact that all in one adhesive requires ionizing medium that is represented in water to start the reaction. Water could be trapped in the hybrid layer that’s the reason why it required cautious air blowing following the adhesive application. While there was no statistical difference between both substrates whether wet or dry using either Activa or RelyX unicem cements.

In the matter of effect of the substrate, it was found that under wet or dry conditions both types of cements (Relyx unicem and Activa) when bonded to enamel, showed statistically higher shear bond strength than with dentin. It was found that Relyx unicem offered high bond strength with enamel due to the chemical adhesion rather than micromechanical bonding which is achieved by the functional monomer (methacrylate phosphoric esters) which chelates calcium ions of hydroxyl-apatite [23]. Conversely, Lee et al. [24] found that enamel showed low bond strength values when bonded to self-adhesive resin cements. De Munk [25] demonstrated that self-adhesive cements characterized with low PH, but it hardly demineralizes tooth surface, because of its high viscosity and poor penetration thus resulting in low bond strength.

In regard to modes of failure using SEM, RelyX and Activa bioactive cements, both, showed mixed and adhesive mode of failure. This indicates that bioactive and self-adhesive cement interacted superficially with minimal penetration in the tooth substrate. Regarding dentin samples, the prevalent adhesive failure could be attributed to the high-water content of dentin that increases water fraction and interferes with complete polarization of the adhesive. Partial obliteration of dentinal tubules with resin plugs revealed in ACTIVA SEM sample (figure 5), may be attributed to the presence of nanoparticles that helped in the flow of cement and formation of hybrid layer. In accordance with the current study, Flurry et al. [26], Lin et al. [27] and Viotti et al. [28] observed mixed and adhesive pattern of failure indicating that no real chemical bond was established.

Despite the modest performance of ACTIVA bioactive cement, it showed comparable results to the well reputed self-adhesive resin cement RelyX. This performance could be attributed to its various setting mechanism; light and chemical activated polymerization and acid-base reaction that involve ionic attraction of two carboxyl groups in the cement to calcium in tooth substrate. The bioactive cement has an ionic resin network and bioactive nanoparticles that fill the gaps in the matrix and minimally penetrate the dentinal tubules that supports ionic bonding and eventually extends the longevity of the restoration [5]. Future clinical long term studies are required to test the material clinical behavior and longevity and to assess the effect of pH fluctuation, load switches and enzymatic challenges on the bonding joint.

5. Conclusion
Within the parameters used and the limitations of this study, the following could be concluded: Both tested cements exhibited comparable shear bond strength to either wet or dry tooth substrate (enamel or dentin), with superiority of self-adhesive resin cement RelyX. Enamel substrate showed improved bond strength than dentin regardless wet or dry conditions and cement type. RelyX and Activa Bioactive cements, showed mixed and adhesive mode of failure

Clinical Relevance
Despite the fact that the resin cement outperformed the bioactive cement, the new bioactive cement containing nano particles showed comparable results and is considered as an advantageous candidate for bonding procedures in the clinic especially when moisture control is unattainable.

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