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Mechanical Stability of Self-Adhesive/Ion-Releasing Resin Composites

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Abstract: The purpose of this study was to assess the effects of water storage on the surface micro-hardness (VHN) and fracture toughness (K_{IC}) of two self-adhesive restorative materials compared to traditional resin composite and resin-modified glass ionomer cement (RMGIC) restorative materials. Methods: Two self-adhesive materials (Activa and Vertise Flow), a nonflowable composite (Filtek Z250), and an RMGIC (Fuji II) were evaluated. Hardness measurements ($n = 12$) were recorded at three time intervals: (i) one-hour post-irradiation; (ii) after one day of storage in water at 37 °C; and (iii) after 90 days of storage in water at 37 °C. Fracture toughness (K_{IC}) measurements ($n = 12$) were conducted after one day of storage in water at 37 °C and 90 days of storage in water at 37 °C. ANOVA and Tukey post hoc tests were used for statistical analysis. Results: Baseline VHN data were 38.2–58.3, decreasing significantly to 28.8–55.6 following 90 days of water storage. The Filtek Z250 had the highest VHN before and after storage, while the Activa had the lowest. K_{IC} values varied between 0.98–1.32 MPa·m^{0.5}. The highest value was for the Filtek Z250 while the Fuji II showed the lowest value (after both 1 and 90 days of storage in water). However, K_{IC} values decreased significantly after storage, except for the Fuji II. Conclusion: Self-adhesive/ion-releasing resin composites were negatively affected by water storage. Material reinforcements are possible future areas to explore.

Keywords: self-adhesive; ion-releasing; resin composites



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1. Introduction

The current first-choice material for restoring anterior and posterior teeth is resin-based composites (RBC). Their popularity soared when dental amalgams were phased out after the Minamata Convention on Mercury, aided by the fact RBCs are aesthetically pleasing and have similar clinical success rates to amalgam [1,2]. Although RBCs have undergone several advancements to improve their performance and are commonly used, their longevity is still negatively affected by key shortcomings [3,4]. The main reported reasons for RBC failure are (and have been for several decades) bulk fracture and recurrent caries [4–6].

RBCs for posterior teeth and multi-surface restorations face particularly challenging environmental and mechanical challenges [7]. Enzymes, water, and cariogenic acids found

in the oral cavity contribute to RBC degradation and hydrolysis [8]. These factors can negatively affect the material's microhardness and resistance to wear and fracture, thus reducing its clinical longevity [8–10]. RBCs are likely to fail if the bonding interface deteriorates and/or secondary caries form [10]. One method of preventing this is by adding reactive particles to the composite material. This can assist in two ways, (i) encourage the damaged tooth to remineralize, and (ii) perform an antibacterial function by neutralizing the effects of acid-producing micro-organisms.

One class of material that is attracting much interest in the field of dentistry is self-adhesive bioactive restorative materials. In this field, “bioactive” is used to refer to a material that can form an appetite-like phase when immersed in a physiological-like solution [11]. The first materials of this type employed in dentistry were resin-modified glass ionomer cement (RMGIC) and glass ionomer cement (GIC). They can reduce the incidence of caries by releasing and recharging fluoride ions [12–14]. However, these materials do have a major drawback as their mechanical properties are poor compared to RBCs [13]. They, therefore, do not withstand occlusal loading well, and they fail too often to be considered as a material for many final direct restorations [15]. Bioceramic particles have been incorporated into dental materials to prevent recurrent carious lesions and simultaneously promote remineralization of tooth structure. Examples include calcium phosphate, amorphous calcium phosphate, and bioactive glasses [16]. However, doubts have been raised over their mechanical and physical performances as the ions released could create voids inside the dental material [17].

Recently, two new self-adhesive resin composites have been introduced, including one that is marketed as bioactive. These are ACTIVA™ Bioactive Restorative (Pulpdent, Watertown, MA, USA) (ACT) and Vertise Flow (Kerr Dental, Brea, CA, USA) (VRF). ACT is described by Pulpdent as a ‘bioactive composite’ and although the company maintains that it releases a greater amount of fluoride than GIC [18], Garouchi et al. [19] have questioned this claim. ACT has been described by some as a reinforced version of RMGIC because their chemical setting reactions are the same and they both include a modified polyacid with a small amount of water [20]. In addition, bioactive fillers and a bioactive matrix developed by Pulpdent are incorporated into ACT [19]. VRF on the other hand is not bioactive but is self-adhesive. Ion-releasing materials may be particularly useful for individuals with high caries risk, promoting caries reversion and bonding to lesions with little to no enamel structure [20].

Self-adhesive restorative composites were first marketed in 2009. They share some characteristics with self-etch adhesive systems, e.g., the fact that both employ mild acidic monomers such as ultra-mild carboxylic methacrylates (i.e., 4-MET, pH = 3–4), glycerol phosphate dimethacrylates (GPDM, pH = 1.9), or phosphate ethyl methacrylates (BMEP) [12]. These monomers create a hybrid layer by partially etching the surface of the restored tooth [13]. The level of adhesion between the tooth and the restorative material thus relies on the degree of tooth-surface decalcification achieved by the acidic monomers. Self-adhesive materials reduce the potential risk of postoperative sensitivity, simplified usage steps, and minimized time for possible blood or saliva contamination [12].

The strength of RBCs can be assessed through a fracture toughness (K_{IC}) test, as the existence of a notch is considered a reliable indicator of surface defects [7]. Furthermore, fracture toughness is the only property found to be strongly associated with the clinical fracture of RBCs [14]. It is therefore a parameter of prime importance for the success of direct restorative materials in the long term in clinical practice.

The use of various accelerated aging methods, such as water storage, can give some indication of a material's durability [10]. Water molecules are absorbed into the matrix-filler interface leading to the hydrolytic breakdown of the bonds [15]. This alteration of the chemical structure and the structural stability takes effect over a long period of water exposure [16,17]. Several factors influence the rate of absorption, including the type of monomer, surrounding environment, surrounding temperature, coupling agent used, and the degree of conversion [18–20].

Another significant physical–mechanical characteristic is surface hardness, which can be defined as the measurement of a material’s resistance to indentation under the application of a constant load. Although some results have been contradictory, there does seem to be a positive relationship between wear resistance and the hardness of RBCs, meaning that material with lower hardness is likely to wear faster [21]. Several factors can affect hardness values, such as filler size, degree of conversion, and compressive strength [22]. A higher filler load increases RBC hardness [22], giving rise to the need for the hardness of new materials to be evaluated in vitro.

Since CN and ACT have only been recently introduced, their physical and mechanical properties are yet to be investigated. Comprehensive testing of these materials is important to provide accurate clinical recommendations for their utilization. Therefore, this present study aims to assess the effect of water storage on surface hardness and fracture toughness of two self-adhesive restorative materials and determine how this compares to traditional RBC and RMGIC restorative materials. The tested hypotheses were as follows:

- No difference would be observed in VHN for the ACT, VRF, XTE, and Fuji II (i) compared to every other material, and (ii) after 90 days in water storage compared to 1 day.
- No difference would be observed in K_{IC} for the ACT, VRF, XTE, and Fuji II (i) compared to every other material, and (ii) after 90 days in water storage compared to 1 day.

2. Materials and Methods

The materials investigated are presented in Table 1. Four materials were used: a self-adhesive, bioactive material (ACT), a self-adhesive material (VRF), a traditional resin composite (Filtek Z250, 3M Oral Care, St. Paul, MN, USA), and an RMGIC (Fuji II, GC, Tokyo, Japan). Groups were chosen to represent composites used for different clinical applications with varying percentages of resin and filler content. The traditional resin composite and RMGIC were used as standard materials for comparison with the self-adhesive materials ACT and VRF.

Table 1. Materials tested and their composition.

Material		Manufacturer	Type and Shade	Filler Load		Filler Type	Resin Matrix
Code	Name			Vol%	Wt%		
ACT	Activa	Pulpdent, Watertown, MA, USA	Resin-based ion-releasing material (paste-paste with tip)/A3 shade	N/A	56%	Reactive ionomer glass fillers of bioactive glass and sodium fluoride	Patented ionic resin matrix, shock-absorbing rubberized resin (diurethane and other methacrylates with modified polyacrylic acid 44.6%)
VRF	Vertise Flow	Kerr Dental, Brea, CA, USA	Self-adhering flowable Composite/A3 shade	N/A	70%	Ytterbium fluoride, barium aluminosilicate glass, prepolymerized fillers, and colloidal silica	GPDM adhesive monomer, UDMA, BisGMA, and other methacrylate comonomers, photoinitiators
XTE	Filtek Z250 Universal	3M Oral Care, St Paul, MN, USA	Resin composite nanofilled (syringe)/Body A2	60%	78.5%	Zirconia and silica particles	BisGMA, UDMA, TEGDMA, BisEMA
Fuji II	Fuji II LC	GC, Tokyo, Japan	Polyacrylic acid, 2-HEMA, dimethacrylate	N/A	58%	Al-Si-glass with a Powder/liquid ratio (g/g) = 3.3/1.0	Polyacrylic acid, 2-HEMA, dimethacrylate

UDMA, urethane dimethacrylate; DCP, tricyclodecane-dimethanol dimethacrylate; PEG-400 DMA, polyethylene glycol 400 dimethacrylate; BisGMA, bisphenol A-glycidyl methacrylate; HEMA, 2-hydroxyethyl methacrylate; TEGDMA, tetramethylene glycol dimethacrylate; BisEMA, ethoxylated bisphenol A glycol dimethacrylate; N/A, data not available.

2.1. Surface Hardness

Twelve disk-shaped specimens (8 mm × 2 mm) were prepared following the manufacturer's instructions for each material (see Table 1). Each mold was laid on a transparent Mylar strip, then 1 mm glass slides were positioned on each side and pressed together. Specimens were cured for 20 s with an LED curing unit at 1200 mW/cm² (Elipar S10, 3M Oral Care, Paul, MN, USA). A calibrated radiometer was employed to confirm the irradiance (MARC™ Resin Calibrator, Blue-light Analytics Inc., Halifax, NS, Canada) each time the curing unit was utilized. Specimens were removed from their molds and finished using polishing disks (OptiDisc; Kerr Hawe SA, Bioggio, Switzerland), starting with coarse, medium, and then fine. A handpiece was employed for polishing at 15,000 rpm.

A Vickers microhardness instrument (FM-700, Kawasaki, Kanagawa, Japan) was utilized to measure the surface hardness of each specimen at three time points: (i) at one-hour post-irradiation; (ii) at one day post-irradiation and stored in water at 37 ± 1 °C; and (iii) at 90 days post-irradiation and stored in water at 37 ± 1 °C. The Vickers hardness number (VHN) was obtained with a 300 g load applied for 15 s at 23 ± 1 °C. Three equidistant indentations were made on each specimen, along with 1 mm adjacent indentations from specimen margins for each measurement. Further information on surface hardness may be found elsewhere [23].

2.2. Fracture Toughness

Using a brass mold lined with polytetrafluoroethylene (PTFE) that conformed to British Standard 54479:1978, 12 single-edge notched (SEN) samples were fabricated for each group ($n = 12$). The LED curing unit mentioned previously was used to photopolymerize each sample for 20 s. Specimens were irradiated in six overlapping areas along their length and any excess composite material on the edges of the specimens was trimmed away using 320-grit metallographic paper. Specimens were then stored in small bottles of distilled water and incubated for 24 h at 37 °C. A stereomicroscope (EMZ-5; Meiji Techno Co., Ltd., Saitama, Japan) was used at 1.5× magnification to measure the crack length. Measurements were taken with an accuracy of 0.1 mm. An electronic digital caliper (Powerfix, OWIM GmbH & Co., Neckarsulm, Germany) was employed to take the dimensions of the specimen with an accuracy of 0.01 mm. Measurements of the height and width were taken at the center and two other points of each specimen. A universal testing machine (Zwick/Roell-2020, 2.5 kN load cell) at 23 ± 1 °C was utilized to measure the fracture toughness (K_{IC}) by flexural loading. Each beam specimen was subjected to a central load in a three-point bending mode, at a crosshead speed of 1.0 mm/s until fracture. Further information on fracture toughness may be found elsewhere [24].

The following formula was used to calculate the fracture toughness from the load values:

$$K_{IC} = \left[\frac{PL}{BW^{1.5}} \right] Y \quad (1)$$

P = Load at Fracture	B = Thickness of the Specimen
L = distance between the supports	Y = calibration function for a given geometry
W = width of the specimen	a = notch length
$Y = [2.9 (a/w)^{1/2} - 4.6 (a/w)^{3/2} + 21.8 (a/w)^{5/2} - 37.6 (a/w)^{7/2} + 38.7 (a/w)^{9/2}]$	

Equation (1): Fracture toughness equation.

2.3. Statistical Analysis

Data for all groups were collected and analyzed using SPSS 22.0 (IBM SPSS Statistics, SPSS Inc., Armonk, NY, USA). The Shapiro–Wilk test was implemented to confirm the normality of the data. Levene's test of homogeneity also confirmed the equality of variance. For microhardness, one-way and two-way analysis of variance (ANOVA) with a Tukey post hoc test ($\alpha = 0.05$) were performed to identify differences in hardness (dependent) between different materials and time (independent variable). For K_{IC} , one-way and two-

way ANOVA with a Tukey post hoc test ($\alpha = 0.05$) were performed to identify differences in K_{IC} (dependent variable) between different materials and time (independent variable). Each group was considered independent and thus repeated measurement ANOVAs were not used.

3. Results

Table 2 presents the VHN data for each type of material and the data are graphically shown in Figure 1. The VHN baseline data after 1 h of dry storage ranged between 38.2 and 58.3, which then decreased to a range between 28.8 and 55.6 after 90 days of water storage. XTE had the highest VHN before and after storage, followed by Fuji II. ACT had the lowest VHN. The aging period caused a significant reduction in VHN over time; the baseline values (after 1 h) were higher than those recorded at 90 days ($p < 0.05$). This decrease over time in VHN varied from 5% (XTE) to 25% (ACT). On the other hand, the surface hardness increased for ACT ($p < 0.05$) after being stored for 24 h in water; the others showed no significant differences ($p > 0.05$).

Table 2. VHN values (mean and standard deviation) for each type of material with respect to storage time.

Materials	Dry		Distilled Water	
	1 h	1 d	90 d	Change %
ACT	38.2 (1.0) ^{a,2}	43.1 (1.2) ^{b,2}	28.8 (1.2) ^{c,2}	25%
VRF	40.1 (2.0) ^{a,2}	38.1 (1.1) ^{a,3}	30.7 (1.6) ^{b,2}	23%
XTE	58.3 (1.7) ^{a,1}	60.5 (2.2) ^{a,1}	55.6 (1.9) ^{a,1}	5%
Fuji II	48.8 (1.6) ^{a,3}	52.1 (2.2) ^{a,4}	40.5 (1.2) ^{b,3}	17%

The same superscript letters indicate no significant difference ($p > 0.05$) for each material across time points. The same number of superscripts indicates no significant difference ($p > 0.05$) between materials at each time point. $n = 12$ samples for all tests.

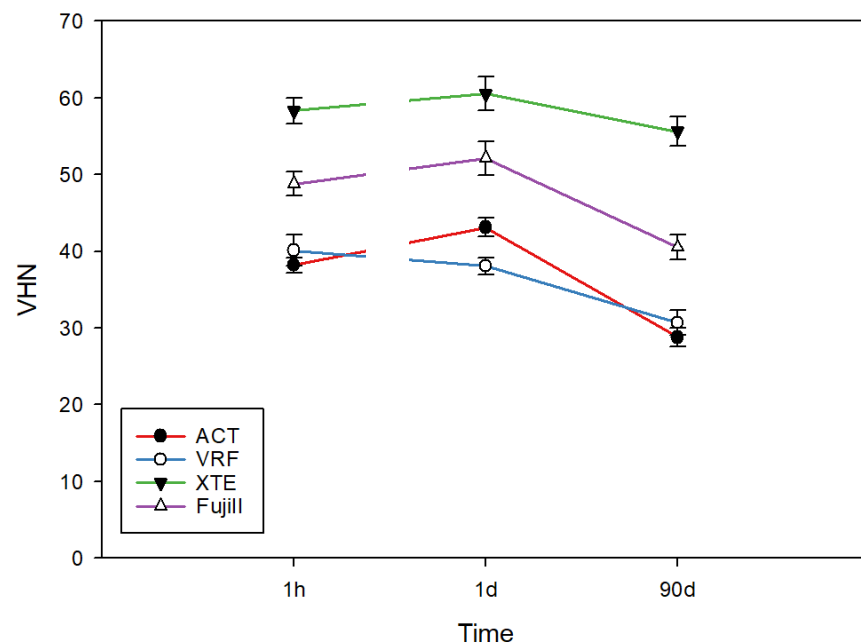


Figure 1. Vickers hardness (VHN) of materials measured at three time intervals: after 1 h dry, after 1 d, and after 90 d at 37 °C. The broken line indicates that the 1 h time point was obtained under dry storage while 1 and 90 d were obtained after underwater storage at 37 °C. $n = 12$ samples for all tests.

Fracture Toughness

Fracture toughness (K_{IC}) values are shown in Table 3 and Figure 2. The K_{IC} values varied between 0.98 and 1.32 $\text{MPa}\cdot\text{m}^{0.5}$. Significant differences were found among materials on day 1 ($p < 0.05$). The highest value was found for XTE, with Fuji II showing the lowest ($p < 0.05$) (after both 1 and 90 d of storage). However, all K_{IC} values decreased significantly with the time spent in storage ($p < 0.05$), except for Fuji II. Ultimately, ACT, VRF, and Fuji II all had comparable K_{IC} results after 90 days of aging ($p > 0.05$).

Table 3. Fracture toughness (mean and standard deviation) after 1 and 90 days of storage in water.

Material	Fracture Toughness K_{IC} ($\text{MPa}\cdot\text{m}^{0.5}$)	
	1 d	90 d
ACT	1.11 (0.09) ^a	0.85 (0.07) ^e
VRF	0.98 (0.08) ^b	0.76 (0.07) ^e
XTE	1.32 (0.07) ^c	1.14 (0.06) ^f
Fuji II	0.79 (0.05) ^d	0.74 (0.07) ^{d,e}

The same superscript letter indicates no significant difference ($p > 0.05$) between materials when compared across both material and day. $n = 12$ samples for all tests.

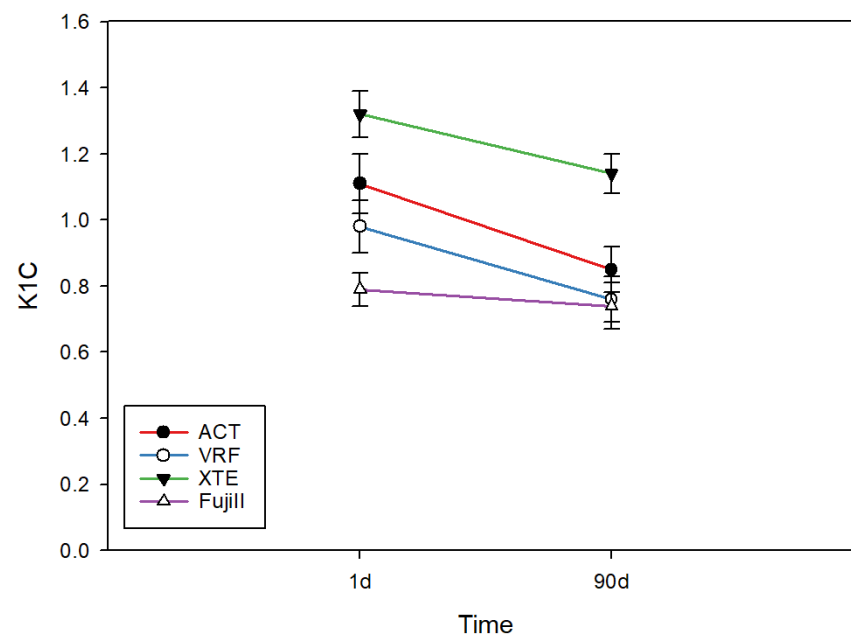


Figure 2. Fracture toughness of materials after 1 and 90 d of storage in water at 37 °C. $n = 12$ samples for all tests.

4. Discussion

The relatively new material category, self-adhesive/ion-releasing resin composites, was designed to interact with their surrounding environment. While this interaction with tooth structure should chemically bond and release ions, they should also withstand the harsh oral environment including occlusal forces, abrasive foods, aqueous environment, rapid thermal changes, bacteria, and enzymes. Such an environment may affect a variety of resin–composite properties, including wear resistance, microhardness, dimensional stability, and fracture resistance [9,16]. Thus, the purpose of this study was to study how water storage affected (i) surface hardness and (ii) fracture toughness of two self-adhesive restorative materials, including one that is bioactive, in comparison to traditional RBC and RMGIC.

Hardness values in this study were from high to low: XTE, Fuji II, VRF, and then ACT. This led to the rejection of the first part of the first hypothesis, which was proportional to filler loading for the resin composites. Filler size, shape, and fraction are all factors known to influence VHN [22]. The diffusion-controlled degradation affected all materials to different magnitudes, with XTE showing a 5% reduction compared to ACT showing a 25% VHN reduction at 90 days. This led to the rejection of the second part of the first hypothesis. Furthermore, the more hydrophilic nature of the monomers used in self-adhesives likely accelerated the surface plasticization, as reflected by the lower VHN of the specimens over a longer period of water storage.

One of the principal reasons for the failure of direct restorative material is bulk fracture [25], which is why it is crucial to measure the fracture toughness of any new proposed or introduced material when conducting an evaluation [26]. In this study, all specimens were fabricated and then tested with the single-edged notched (SEN) beam method [27]. Although this is a widely used method, the construction of the sample, particularly the notch, is challenging. Moreover, as different studies employ different methods for producing the notch, they tend to report varied results [28]. In this study, to ensure consistency and comparability, the preparation and measurement steps were standardized.

The fracture toughness at baseline (24 h) for the materials tested, in descending order, were: XTE, ACT, VRF, and Fuji II LC. This led to the rejection of the second null hypothesis that there would be no difference in the fracture toughness between materials. After 90 days of water storage, there was no statistically significant difference between the K_{IC} of ACT, VRF, and Fuji II. Material stability behavior did vary when compared to the baseline values. The K_{IC} of Fuji II was the lowest at baseline but was not significantly affected by 90 days of water storage.

One likely explanation for ACT's initial high fracture toughness is that it contained a rubberized resin that increased the ability of the material to absorb shocks [29]. In addition, it was observed during the testing of ACT that when the load was applied, the sample had a notable bend before failure. Other researchers have noted this and suggested that this characteristic is due to the material's low flexural modulus [30]. We did not test the elastic modulus in this investigation, but it is known that it affects K_{IC} as the fracture toughness is proportional to the square root of the product of the elastic modulus and the fracture energy [31]. A material with a low elastic modulus will show greater distortion, which is not a favorable characteristic for most dental applications. Furthermore, high fracture toughness combined with easy distortion is undesirable as it reduces the restorative material's strength and may lead to uneven occlusal forces being applied to the material. The occlusal pressure exerts a high tensile load on the material's surface, reducing adhesion to the tooth structure. This could ultimately cause the tooth surface to break as a result of occlusal load on the restorative materials causing lateral expansion deeper into the preparation.

The samples were stored in water for 90 days before their fracture toughness was assessed in this study. Being stored in water, especially for long periods, can lead to water intake and any cracks within the material may expand, thus altering the properties of the material. Our study showed that the Fuji II LC produced the lowest fracture toughness. However, water storage did not significantly affect the K_{IC} , therefore this was the most stable material studied. The fact that the Fuji II LC is quite resistant to water-induced deterioration has been reported in previous studies about RMGIC; this property has been ascribed to the formation of a more robust polysalt matrix after water storage [32,33].

Even though ACT contains water and acid, its K_{IC} value decreased the most after storage in water for 90 days. This could be attributed to the fundamental differences between the ACT and Fuji II, which could negatively affect the material's stability. For example, the presence of a modified polyacrylic acid in ACT might prevent the formation of an acid that is stable in water. For this reason, it is recommended that restorative materials are tested for their fatigue performance after they have been stored in water for some time [34]. Thus, testing after storage/aging is important to more accurately reflect clinical

performance after material placement. Water storage is a relatively easy and inexpensive method to achieve this.

The VRF self-adhesive resin composite showed generally similar behavior to ACT. However, the difference in monomer composition and filler load resulted in higher initial hardness values. The higher (70% vs. 56% vol) filler load of the VRF may result in higher initial viscoelastic properties [35]. Furthermore, Bis-GMA-loaded materials have shown good mechanical properties when optimally polymerized [36]. Even though Bis-GMA and UDMA incorporated in VRF are known to be susceptible to hydrolytic degradation, the GPDM monomer (used to promote self-adhesion) has previously been shown to be relatively hydrophilic. Indeed, GPDM is a functional monomer used in the gold standard etch and rinse adhesive system (Optibond FL, Kerr, FL, USA). Thus, the high solubility of GPDM likely caused the significant reduction in surface hardness as well as the K_{IC} [37].

Fiber reinforcement has been proposed by some scholars as a way of enhancing the mechanical performance of restorative materials that release ions [38,39]. This is relevant given the relatively inferior performance of VRF and ACT herein. For example, studies have reported significant improvements in mechanical properties—almost 2 to 3-fold—after the addition of short glass fibers to the RMGI powder [38–40]. The addition of short glass fibers improves fracture toughness because of the higher resistance to crack propagation as well as the mechanical stability that was improved by the fibrous structure. However, the physical properties and the ion-releasing capability of such fiber-reinforced-ion-releasing materials are yet to be studied. Other materials with future applications in dentistry are piezoelectric materials [41].

5. Conclusions

Within the limitations of this study, the self-adhesive materials ACT and VRF showed inferior microhardness (VHN) and fracture toughness (K_{IC}) compared to the nonflowable resin composite XTE. Fuji II showed higher VHN results but lower K_{IC} . The VHN of all materials, except XTE, significantly reduced following 90 days of water storage. The K_{IC} of all materials, except Fuji II, was significantly reduced following 90 days of water storage. Overall, water storage is recommended in the testing of restorative materials to gain more clinically relevant information.

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