BJOS
Brazilian Journal of Oral Sciences

Original Article

Volume 21
2022
e225263

Comparative of flexural strength, hardness, and fluoride release of two bioactive restorative materials with RMGI and composite resin

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Editor: Altair A. Del Bel Cury

Received: April 11, 2021
Accepted: October 24, 2021

Aim: This study was fulfilled to evaluate the flexural strength, micro-hardness, and release of two fluoride ions of bioactive restorative materials (Cention N and Activa Bioactive), a resin modified glass ionomer (Fuji II LC), and a resin composite (Filtek z250). **Methods:** Forty samples from four restorative materials (Activa Bioactive, Fuji II LC, Cention N, and Filtek Z250) were provided according to the current standards of ISO 4049/2000 guide lines. Subsequently, the samples were stored for 24 hours and 6 months in artificial saliva, and successively, flexural strength and micro-hardness of the samples were measured. For each studied groups the pH was decreased from 6.8 to 4 in storage solution. The rate of changes in fluoride ion release was measured after three different storage periods of 24 hours, 48 hours, and 6 months in distilled water, according to the previous studies' method. Two-way ANOVA, One-way ANOVA, Tukey HSD Pair wise comparisons, and independent t-tests were used to analyze data (α= 0.05). **Results:** The highest flexural strength and surface micro-hardness after 24 hours and also after 6 month were observed for Cention N($p<0.001$).Flexural strength of all samples stored for 6 months was significantly lower than the samples stored for 24 hours($p<0.001$). The accumulative amount of the released fluoride ion in RMGI, after six-month storage period in distilled water was considerably higher ($p<0.001$) than 24 hours and 48 hours storage. The amount of fluoride ion release with increasing acidity of the environment (from pH 6.8 to 4) in Fuji II LC glass ionomer was higher than the bioactive materials ($p<0.05$). **Conclusion:** The flexural strength of RMGI was increased after storage against the Activa Bioactive,Cention N and Z250 composite. Storage of restorative materials in artificial saliva leads to a significant reduction in micro hardness. The behavior and amount of released fluoride ions in these restorative materials, which are stored in an acidic environment, were dependent on the type of restorative material.

Keywords: Materials testing. Physical phenomena. Dental materials. Saliva, artificial.

Introduction

Nowadays, the application of resin-based restorative materials is getting increased in Dentistry due to esthetic issues, ease of application, and the capability of chemically bonding to dental structures[1].

Different types of direct esthetic restorative materials are available to dentists, including composites and glass ionomer cements[2]. Formulation improvement, leading to the increase in durability of these restorative materials, and made them as the preferable materials for dentists[3].

Despite the existence of these benefits, there are some problems related to usage of these materials. Marginal integrity, polymerization shrinkage, secondary caries, and post-operative hypersensitivity are recognized as the problems in usage of these substances in the practical work[4].

Glass-ionomer cements are one of the most useful dental materials used in restorative dentistry due to their properties such as the ability of fluoride release, the intrinsic adhesion to the dental structures, the coefficient of thermal expansion similar to the dental structures and their biocompatibility[5]. Despite these benefits, glass ionomers have some limitations such as high wear, solubility, poor mechanical properties, and low strength against occlusal forces[6,7].

Advancement in the science of dental materials leaded to the introduction of bioactive restorative materials in recent years[8]. These materials present a combination of benefits for using glass ionomers, resin modified glass ionomers and composites. Bioactive products actively participate in ion-exchange cycles and help to maintain dental structures and oral health[9].

These materials react to the changes in oral cavity environment to produce useful changes in salivary, dental, and restorative properties. This issue is introduced as "Smart" behavior in this type of restorative materials[10]. Most of the bioactive materials, in addition to their optical and chemical polymerization capabilities, contain polyacid components and glass particles, which affect the reaction of acid-based hardening and therefore include three step hardening mechanisms[11].

Activa Bioactive restorative material was introduced in 2013 by Pulpdent Company. It was reported that, this restorative material resemblance to composite resin, is durable and resistant to abrasion wear, and also can stimulate remineralization and apatite formation[9]. This process can lead to a greater adaptation and marginal seal at the edge of restorations and can ultimately reduce microleakage and secondary caries[12,13].

Activa lacks Bis-phenol A, Bis-GMA, and BPA; therefore, biocompatibility of the material is higher than ordinary composites[14]. It was reported that, this restorative material reacts to continuous changes in pH in oral environment to help the reinforcement and recharge of ionic properties of saliva, tooth, and the substance itself[11,15,16].

Cention N is restorative substance belongs to a new group of materials known as alkazite. There is a resin based tooth-colored material, which has a self-curing set-

ting mechanism with selective light curing capability. It is used as Bulk-Fill to repair teeth too[17]. Fundamentally, alkazites are able to release acid neutralizing ions due to their alkaline fillers; and hence, Cention N is able to release calcium, fluoride, and hydroxide ions in to the oral environment[18]. It was reported that, some mechanical and chemical properties of bioactive restorative materials were lower, compared to restorative composites[19,20].

It was stated that, the release of ions from bioactive materials could lead to the establishment of micro cracking and reduction in mechanical properties[21]. There are few studies that have evaluated and compared the physical and mechanical properties of these materials with the two groups of accepted restorative materials in usual services in dental clinic (composites and glass ionomer cements). On the other hand, in these few studies, the properties of materials in medium or long term storage in the similar conditions of the oral cavity environment have not been studied[19].

The aim of this study was to compare the flexural strength, microhardness, and release of fluoride ion in two bioactive materials and resin modified glass ionomer cement with conventional composite after a six-month storage period in aqueous and acidic environments. The null hypothesis is the flexural strength, micro-hardness and release of fluoride ions of Cention N and Activa Bioactive have no different with resin modified glass ionomer (Fuji II LC) and a resin composite (Filtek z250).

Materials and Methods

This laboratory-experimental study evaluated four direct restorative materials as follows:

Filtek Z 250 (3M ESPE, St Paul, MN, USA), Fuji II LC (GC Corporation, Tokyo, Japan), Activa Bioactive (Pulpdent Corporation, Watertown, MA, USA), Cention N (Ivoclar Vivadent, Liechtenstein, Switzerland) to measure the properties of flexural strength, microhardness, and release of fluoride ions (Table 1).

Table 1. Investigated materials in the study

Materials	Manufacturer	Composition details
Activa Bioactive	Pulpdent, Corporation, Watertown, MA, USA	Mix of diuretane and other methacrylates with the modified poly acrylic acid (44.6 %), reactive glass filler (21.8 wt.%), Inorganic filler (56 wt.%),patented rubberized resin(Embrace), water.
Cention N	Ivoclar – Vivadent, Liechtenstein, Switzerland	Powder: Inorganic fillers (Ba-Al-Ca-Ba-F silicate glass, Ca-F-silicate glass and customized fillers. Liquid: Urethane dimethacrylate, triclodecan-dimethanol dimethacrylate, poly ethyleneglycol dimethacrylate.
Filtek Z250	3M ESPE, St Paul, MN, USA	BisGMA, UDMA, BisEMA with small amount of TEGDMA, Filler 60%vol silanized zirconia/silica particle.
Fuji II LC (Capsulated)	GC corporation, Tokyo, Japan	Powder: Alumino-fluorosilicate glass, Liquid: 35%HEMA, 25% distilled water, 24%polyacrylic acid, 6%tartaric acid and 0.10% camphorquinone, Bis-GMA and traces of TEGDMA.

Depending on the type of test to determine the characteristics, suitable samples were prepared from four restorative materials.

Assessment of flexural strength

For each one of the restorative materials, 10 bar-shaped samples were prepared using metal molds with dimensions of 2*2*25 mm, in terms of the ISO 4049/2000 guidelines. Each restorative material was prepared in terms of the manufacturer's instructions and placed in metal molds. One transparent celluloid strip was placed on the samples' surface, and light-cured (Drs Light AT, Good Doctors co., Ltd, Incheon, Korea) with an intensity of 1200 mw/cm^2 for 20 seconds over a glass slide. The intensity of the device was assessed by a radiometer (Demetron L.E.D. Radiometer, SDS/Kerr) and curing was performed based on the length of samples, as overlapping of the cured areas at 4 regions for each one of the samples. After removing the samples from metal molds, they were randomly divided into four groups. So, each group included two subgroups involved five samples (n=5) to assess their flexural strength at 24 hours and 6 months (in total, eight subgroups) (chart 1).

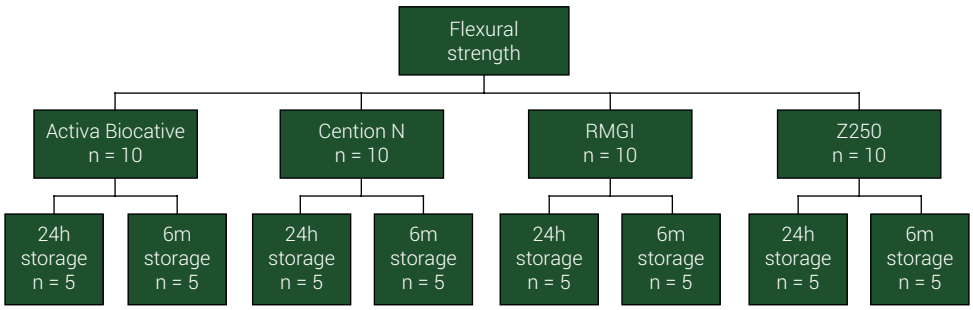

Chart 1. How to distribute samples for assessment of flexural strength.

Artificial saliva with pH=7 was prepared, and the samples were stored in artificial saliva. The composition of artificial saliva included the followings: 7.0 mmol/l $CaCl_2$, 2.0 mmol/l $MgCl_2$, $6H_2O$, 4 mmol/l KH_2Po_4, 30 mmol/l KCl, and 20 mmol/l HEPES buffer, and pH=7[22].

The samples in first group, after being removed from the artificial saliva and rinsed with water, were dried by a paper towel and placed in a mechanical jig (Universal Testing Machine, Zwick/ Roell z020, GmbH Co, Germany) to test the three-point bending .

The speed of applied force was 0.5 mm/min, and the span in between supports was 20 mm. Flexural strength valueswere calculated according to the following formula in terms of mega Pascal's: ∂=3 Fl / 2 b d^2

Flexural strength values of the second group of samples were assessed after six-month storage in artificial saliva according to the above-mentioned method.

Data were analyzed by Two-way ANOVA and One-way ANOVA for subgroups of 48-hour and 24-hour. Tukey HSD pairwise comparisons was performed (α= 0.05).

Assessment of surface micro-hardness

Ten disk-shaped samples with a diameter of 6 mm and a thickness of 2 mm were prepared for each material (totally 40 samples) by using metal molds. After placing the materials in the molds, celluloid strip was placed on the bottom and surface of samples, and the samples were cured beyond the glass slide for 20 seconds.

The bottom surface of the samples were marked with sharp tip of scalpel, and the superficial surface was polished with 800, 1000, 1200, 1500, 2000, 2500 and 3000 grit sandpaper with reciprocating motions along with water stream, and then were washed by distilled water. The samples were randomly divided into two subgroups to assess 24-hr and 6-month Vickers hardness (8 subgroups) and there were5 samples in each sub-group (n=5).Artificial saliva with pH=7 was prepared and samples were placed in artificial saliva. Surface micro-hardness was assessed using Vickers hardness test by diamond indenter with apex angle of 136°. After 24-hour each of the samples of subgroups 1 to 4 dried and the surface hardness was determined by a surface hardness tester system (ZHVμ, Zwick / Roell, Zwick, GMBH, Germany) in three areas with a distance of at least 300 microns with a force of 100g and a standstill of 15 seconds. The mean values of three regions were recorded as superficial micro-hardness of each sample in kg/mm^2.

In the second group, surface micro-hardness was assessed after a six-month period of storage in saliva for the subgroups 5-8 in terms of the above-mentioned method.

Data were analyzed by two-way ANOVA and one-way ANOVA for 48-hour and 24-hour subgroups and Tukey HSD Pairwise comparison was performed ($\alpha = 0.05$).

Assessment of fluoride ion released before and after increasing, the acidity of the sample storage environment was performed as follows:

Eighteen disk-shaped samples were prepared using metal molds with a diameter of 6 mm and a thickness of 2 mm from each one of the study materials (Fig1.). After the placement of materials in the molds, celluloid strip was placed on the bottom and surface of the samples, and after placing a glass slab on the molds, the samples were

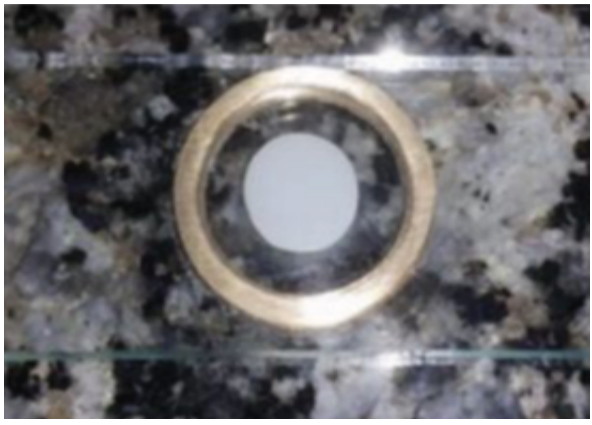

Figure 1. Preparation of disk shaped samples.

cured beyond glass slide for 20 seconds. Both surfaces of samples were polished using sandpaper of 600,800, and 1000 grit with the water stream, and were washed by distilled water. Sample distribution method is specified in Chart-2.

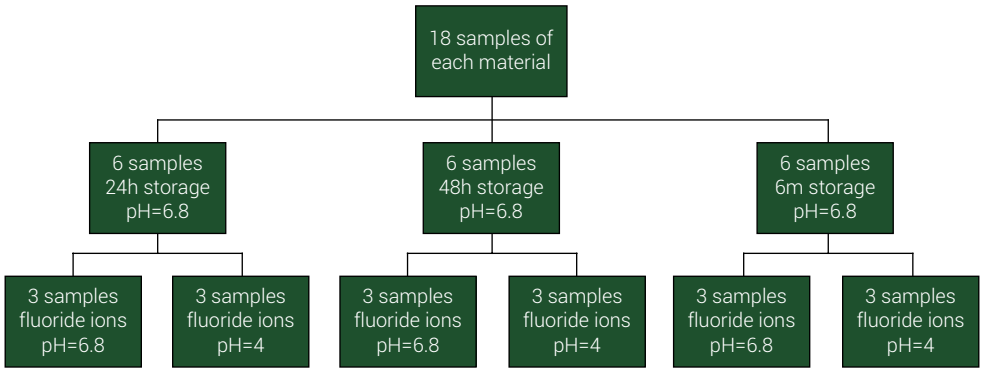

Chart 2. Sample distribution for determining fluoride ion release.

Three of the six study samples (Cen 24 ph=6.8) in group 1 were separately placed for 24 hours in a plastic screw-top container containing 5 ml of distilled water (37°C) with a pH of 6.8. The value of pH was measured directly for each solution by pH-meter system (Metrohm 744, Metrohm Ltd, Herisa, Switzerland). Then one ml of solution of each plastic container was picked using micropipette and was diluted in 9 ml of distilled water. Two ml of fluoride reagent solution (Cat 21060-69) was added to solution and then was properly shaken for 20-30 seconds to achieve a homogeny solution. The prepared solution was placed in the spectrophotometer (DR-5000, HACH Co, Loveland, USA) and the amount of the related fluoride was recorded in mg/l.

The other three samples(Cen 24 pH=4) in group 1 were individually placed for 24 hours in plastic packs containing 5 ml distilled water at 37°C with PH=6.8. Afterward, using 50 ml/mol lactic acid, the PH of solutions reached 4 and the samples were kept for 1 hour[23]. One ml of solution was picked from the each plastic container and was diluted in 9 ml distilled water. The amount of fluoride ions was recorded in mg/l in terms of the above-described method. Therefore, changes in the amount of fluoride ion released from the samples after one hour of reaction with acidic environment and pH reduction from 6.8 to 4 were calculated in one hour with repeated measures at time points of 0, 10, 20, 30, 40, 50, and 60 minutes. In order to make the obtained results similar to clinical situation and determining the effect of sample free surface area on amount of releasing ion from the materials, the data were recorded using the following equation in $\mu g/cm^2$ [24].

$$\text{Ion release} = \frac{\mu g}{m^2} \times \text{Volume of Solution/Area of Sample (Surface area } (cm^2) = 2\pi r\,(r+h)$$

In group 2 and 3 the measurement was performed similar to group 1, except the storage was done in distilled water for 48 hours and 6 months.

This method was also used for three other materials and the changes for ion released from the samples were calculated before and after being placed in the acidic environment at three time points of 24-hr, 48-hr and six months storage.

Statistical analysis

Two-way ANOVA, One-way ANOVA, Tukey HSD pairwise comparisons, and independent t-tests were used to analyze data of ion release among different materials and subgroups (α= 0.05).

Results

Two-way ANOVA showed that the type of restorative material and storage time in artificial saliva had a significant effect on the amount of flexural strength value of restorative materials (respectively, p=0.001 and p=0.001). In addition, the interaction between storage time and type of restorative material factors was significant (p = 0.006). Therefore, for 48-hour and 24-hour subgroups analysis used to compare the type of restorative materials with one-way ANOVA and Tukey's post hoc tests.

Table 2 presents the statistical indices of flexural strength for 4 study materials at two timepoints of 48-hour and 24-hour measurement. The lowest flexural strength was for RMGI (Fuji II LC) and the highest one was for Cention N at two time points of 24-hr and 6-month storage.

Table 2. Mean flexural strength values of study materials at two storage timepoints in artificial saliva.

Material	24 hours	*P-Value	6 months	*P-Value
Activa Bioactive	111.82±1.28 **A**	0.0001	90.11±0.94 **A**	0.0001
Cention N	130.41±3.11 **B**		101.61±1.53 **B**	
RMGI	26.71±2.12 **C**		40.55±1.43 **C**	
Z_{250}	100.68±1.83 **D**		70.58±2.61 D	

* One way-ANOVA, Values with different capital letters in each column show a significant difference according to Tukey tests (p <0.05)

Comparison of the materials with Tukey HSD tests showed a significant difference in the amount of flexural strength between all the studied materials and at both storage timepoints of 24 hours and 6 months (P <0.005).

Due to the results, except for Fuji II LC that showed an increase in flexural strength after 6-month storage in artificial saliva (p=0.001), the flexural strength in other groups after 6 month was lower than 24-hr storage.

The micro-hardness tests showed that the minimum surface hardness was observed for Fuji II LC and the maximum value was for Cention N by passing after 24 hours and six months storage in artificial saliva (p=0.001 and p=0.001, respectively)

Two Way ANOVA showed that the type of restorative material and storage time in artificial saliva had a significant effect on surface microhardness value of restorative materials (respectively, p=0.001 and p=0.001). In addition, the interaction between storage time and type of restorative material factors was significant (p = 0.001). Therefore, one-way ANOVA and Tukey post hoc tests were used to compare the effect of restorative materials in 24-hour and 6- month subgroups (Table 3).

Independent T-tests show that except for Cention N, which had no statistical significant difference at two timepoint of storage in artificial saliva (p=0.083), the surface hardness of other materials, after 6 month was considerably less compared to 24-h storage time (p<0.01). In addition, it was identified that, after 24 hours, there was no significant difference in mean value of micro-hardness between two materials of Activa Bioactive and Z250 (P = 0.284), and also between Activa Bioactive, Cention N and Z250 (P = 0.748), while comparison among other groups showed statistically significant difference (p<0.001). There was a significant difference among all groups during 6 months for surface micro-hardness (p<0.001).

Table 3. Mean micro-hardness (kg/mm^2) values of the study materials at two time- points of storage in artificial saliva

Material	24 hours	*P-Value	6 months	*P-value
Activa Bioactive	58.73±1.03 **a**	0.0001	40.84±2.38 **d**	0.0001
Cention N	62.19±1.95 **cb**		59.03±2.97 **cb**	
RMGI	40.91±2.52 **d**		36.78±1.27 **e**	
Z_{250}	60.97±1.83 **ab**		54.96±1.16 **f**	

* One Way ANOVA, Similar Lower Cases letters show no difference, which was achieved by Post HOC Tukey HSD tests(P>0.05)

Two way analysis of variance showed that the main effect of two factors, type of restorative material and storage time in artificial saliva, had a significant effect on the release of fluoride ions from restorative materials (p = 0.001 and p = 0.001, respectively). Also, the interaction between storage time and type of restorative factors was significant (p = 0.000). In other words, the release rate of fluoride ions for different restorative materials at different times of 24, 48 hours and 6 months of storage in artificial saliva were different significantly. Therefore, one-way ANOVA and Tukey post hoc tests were used to compare the effect of restorative materials on ion release in subgroups of 48-hour, 24-hour and 6-month after storage in artificial saliva. Results of fluoride ion release in table 4 and chart 3 show that, storage at three time-points of 24-hr, 48-hr, and 6-month after changes in pH of storage environment from 6.8 to 4, resulted in the significant differences in all the study materials of Fuji II LC, Cention N, Activa Bioactive (P=0.001, P=0.001, and P=0.001, respectively).

At three timepoints of storage in distilled water at 24-hr, 48-hr, and 6-month, and with a decline in pH of the storage medium from 6.8 to 4, the highest rate of changes in fluoride ions release was for Fuji II LC(p<0.001), which was followed by Cention N and Activa Bioactive, respectively (p<0.05).

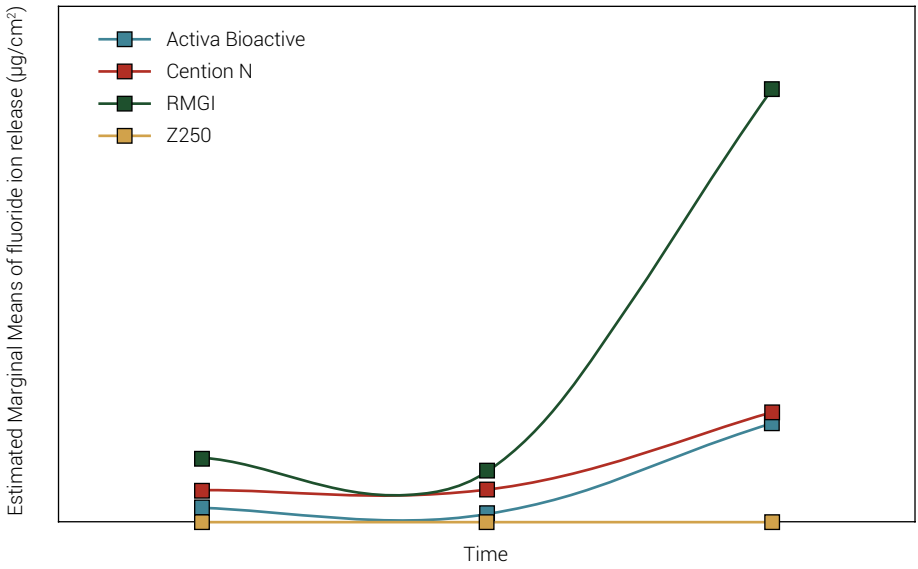

Chart 3. Comparison of the mean values change in fluoride ion release (µg/cm²) from the stored restorative materials following pH reduction from 6.8 to 4 at different time-points.

Also, the highest rate of fluoride ions release with a decline in pH of the environment for two materials of Activa Bioactive and Fuji II LC was at 6-month storage and the least rate was at 48-hr (P=0.001 and P=0.009, respectively).

The rate of changes in fluoride ions release at 24 hours was between these two time-points (p=0.001). In Cention N restorative material, the highest rate of change in fluoride ions release was observed at 6-months, and the storage times at 24 and 48 h had no statistically significant difference (p=0.747).

Table 4. Mean change values of fluoride ion release (µg/cm²) of the study materials after reduce pH of storage solution from 6.8 to 4 at three time points.

Material \ Storage	24 hours	P-Value*	48 hours	P-value*	6 months	P-value*
Activa Bioactive	5.57±0.23	0.0001	3.07±0.46	0.0001	38.82±1.03	0.0001
Cention N	12.72±0.20 a		13±0.64 a		42.88±0.43	
RMGI	24.89±0.05		20.1±0.25		168.32±1.05	
Z_{250}	0		0		0	

* One Way-ANOVA ,Only the same Lower Cases letters show no difference, which was achieved using POH Tukey HSD tests(P>0.05)

Discussion

Being aware of the mechanical and physical efficacies of various materials in dentistry can provide better treatment for the patients[13]. Continuous progresses in the science of materials lead to introducing bioactive restorative materials, which it was claimed that, they have better properties such as apatite construction, stimulating

remineralization, ions release, and control of environment acidity compared to other usual restorative materials[12,13].

However, there are few studies conducted on the effect of time on the properties of bioactive materials. The current study compares some mechanical and physical properties of two tooth-colored bioactive restorative materials, with resin composite, and resin modified glass ionomer cement after 24 hours and 6 months storage in artificial saliva.

The results of study showed that, Cention N has the highest flexural strength at both timepoints of 24-hr and 6-month storage in artificial saliva, followed by Activa Bioactive, Z250, and glass ionomer Fuji II LC, respectively. A higher flexural strength of Cention N could be attributed to the composition of monomer used (UDMA). The findings obtained after 24-hr storage confirmed previous studies, which reported that, glass ionomer flexural strength is lower than resin composite and Activa Bioactive[11,19,25].

Due to the higher amount of resin matrix lead to increase water sorption and softening the samples, it is also obvious that flexural strength of all groups have decreased after 6-month storage in artificial saliva except Fuji II LC.

Long-term durability of restorative materials depends on their mechanical and physical properties, and flexural strength is known as the best scale of dental material resistance and a good index of durability of materials in clinical applications[26]. It was reported that, minimum flexural strength of 80 MPa required for performing the acceptable clinical application in restorative dentistry[27].

Dental composites and restorative materials containing resin are prone to hydrolytic degradation mediated by the effect of water on matrix and filler interface, and also their matrix softening and weakening resulted by water absorption by resin component[28-30]. This issue can be considered as an explanation for reduction in flexural strength of composite z250, Cention N, and Activa Bioactive after 6-month storage in artificial saliva.

It was reported that, ion release from filler particles can result in separating of matrix and filler, and also by micro-cracks formation in interface of filler-matrix interface[21]. This issue can lead to reduction in flexural strength of two bioactive materials after 6-month storage in artificial saliva.

By the way, reduction in flexural strength of Z250 composite was significantly higher in this study compared to Activa Bioactive. Although there are two mechanisms for reducing flexural strength in the Activa Bioactive over time, one is the softening and weakening of the resin base and the other is the release of ions. On the other hand, continuous acid-base reaction after initial setting of glass ionomer and water absorption, can increase the flexural strength over time. This progress could be related to their dual cure setting reaction. The polymerization of resin starts with light curing but acid base reaction progress slowly until further maturation occurs maximum strength over time[31].

Thereby, this might lead to a decline in flexural strength in Activa Bioactive after storage in artificial saliva, which was lower than Z250 composite. Increase in flexural strength of Fuji II LC in the current study was in line with previous studies

showing that, 6-monthsstorage of RMGI leads to an increase in flexural strength[31]. This improvement in strength is due to continuation in setting reaction, which in addition to polymerization of resin presented in ionomer glass, acid-base reaction also continues until achieving the highest rate of material's strength. Delayed substitution of calcium ions by aluminum ions leads to an increase in crosslink and improvement of flexural strength of glass ionomer materials over time. Further investigations are needed to compare these materials in clinical conditions due to temperature and acidity changes, and the presence of enzymes and salivary proteins in oral environment.

Surface hardness is one of the most important mechanical properties of restorative materials, which provides important information on abrasion and setting characteristics of materials[32]. Surface hardness is affected by various factors such as material matrix, the amount and size of filler particles, and the way of filler distribution.

In this study, surface micro-hardness of the studied materials was measured using Vickers test, which is commonly used for dentistry materials. After 24-hr storage in artificial saliva, minimum surface hardness was observed for Fuji II LC, and no statistically significant difference was observed among restorative materials of Z250, Cention N, and Activa Bioactive. Difference in hardness of composite compared to RMGI confirmed in previous studies[33]. Inconsistent with the current studies, Garoushiin et al.[25] 2018 reported that, the value of surface hardness in Fuji II LC after 24-hr storage in dry environment was higher than Activa Bioactive. Although this study is inconsistent with the current study regarding force, conditions of storage, duration, and surface polishing of material. Resin-modified glass ionomer storage in aqueous environment that is inconsistent with dry environments can lead to softening the it's superficial layers, and finally to reducing surface micro-hardness by absorption of water and releasing some ions such as strontium, calcium, phosphate and fluoride from glass ionomer matrix[25].

According to studies by Valanezhad et al.[34], physical characteristics of materials are increased along with an increase in bioactive glass particles. However, the results of this study show that, except for Cention N in other study restorative materials, 6-month storage leads to a significant decline in superficial hardness. In agreement with this study, most previous studies also reported that, long-term storage of restorative materials in aqueous environment can reduce the surface micro hardness[11,17,33,35].Water absorption in resin matrix leads to an increase in volume and its softening, and finally, to a decline in the micro-hardness[9,25].

In resin modified glass ionomer FujiII LC, despite observing an increase in flexural strength during 6 months storage in artificial saliva, the value of surface hardness was significantly decreased. This reduction may be due to the presence of resin components like HEMA as hydrophilic resin component in RMGI, which can increase water absorption at surface layers and leads to approximate decrease of 50% in Vickers hardness[36].

It has been claimed that the release of certain ions, such as fluoride, can play an important role in reducing the incidence of secondary caries, which is the most important cause of failure in tooth-colored restorations[37,38]. So, in this study, we assessed

ion release in aqueous environment and after acidifying the environment. About 50 years ago, Stephan reported that, demineralization of dental structures could be occurred in long-term by exposure to acidic environment with pH lower than critical limit (pH=5.5), which the highest effect of this process could be observed in the first hour of acidity decline[23]. In the current study, the changes in pH and the release of calcium, phosphate, and fluoride ions were individually assessed after decline in environment acidity from 6.8 to 4 in one hour with repeated measures at time points of 0, 10, 20, 30, 40, 50, and 60 minutes.

This method of assessment was also done after 24 hours, 48 hours, and 6 months of storage in distilled water. Ion resin of bio-mineral material of Activa includes acidic groups of phosphate, which improves the reciprocal effect between glass resin and fillers existed in it, and causes releasing of fluoride ion[39,40] as well as high amounts of phosphate ions[39]. Along with the initiation of ionization process, which depends on water uptake, hydrogen ions were separated from phosphate groups, and were replaced by calcium in dental structure. It was reported that, this ionic interaction links resin matrix to minerals in tooth, and forms a strong complex of resin- hydroxy apatite, and can also be effective on causing marginal edge flooding[41] and a decline in microleakage[17,42,43].

Fluoride release from restorative materials was affected by various factors. RMGI cement (Fuji II LC) due to aqueous base and HEMA monomer and the presence of porosity in its structure, has more water uptake and subsequently more ion release in aqueous environments[20,25]. In this study, similar to most of the previous studies, fluoride ion release was higher at all the timepoints compared to two materials of Activa Bioactive and Cention N[17,20,35,44].

Cention N might be the leading cause of producing porosity and bubble during the combination of components of powder and liquid and having alkaline fillers (calcium fluorosilicate), which leads to more ion release in comparison to Activa Bioactive, as is a paste material with resin base[17,35].

The results show that, fluoride release was higher in materials Fuji II LC and Cention N compared to Activa Bioactive. The reason of this issue might be related to producing bubble and porosity at mixing time of Cention N or Fuji II LC in the form of powder and liquid in comparison to Activa Bioactive as a paste material. These porosities can lead to more release of ions in these restorative materials[45]. Inconsistent with the current study, Naghi et al.[44] in 2018 showed that, the amount of fluoride ion release in material of RMGI (Fuji II LC) and Activa Bioactive at time points of storage after 1, 2, 7, 14, 21, and 28 days in distilled water were not significantly different[44]. In their study, the experimental method was different, and the effect of pH decline in storage medium was not investigated on amount of fluoride ion release.

It was stated that, monthly fluoride release at 200 to 300 $\mu g/cm^2$ is required to prevent demineralization of enamel[46]. In the current study, the rate of fluoride release from materials, specifically Activa Bioactive and Cention N during 6 months storage, was lower than this value; therefore, it is possible that, it does not clinically affect the prevention of demineralization of enamel.

Although, the presence of fluoride ion in remineralization process of teeth is not effective as alone, and calcium, phosphate, and OH^- ions had important role. Therefore, it is necessary to conduct further studies on the effect of these materials, regardless of the type of ions, in reducing decay.

Most of the previous studies stated the rate of ions release in unit of volume[25]. However, it should be noted that reporting of ions release rate per surface unit can provide more accuracy for clinical assessments[45]. Accordingly, in the current study, the results of ions release were reported as surface ratio ($\mu g/cm^2$). It was claimed that, some of the bioactive tooth colored restorative materials such as RMGI, Activa Bioactive, and Cention N have the capability of ion release with pH change of environment. These materials through more release of ions in exposure with acidic conditions lead to a significant increase in environmental pH and subsequently prevention of demineralization process[17,47].

The results showed significant increase in pH value after placement at acidic condition. However, it does not seem that, increase acidity of environment (increase in pH between 0.5-0.6) was not enough to be an obstacle for enamel demineralization process. If slump of the pH was remained under critical limit, demineralization process of tooth structure goes on to occur clinically, despite reduction in its speed[48]. Since pH changes can be very important in limited environment of microbial plaque, clinical research is definitely required in this context.

Due to the novelty of these materials and the lack of sufficient research, further studies are needed to determine whether the release of ions leads to the deposition of minerals in the gap between the teeth and restorations, and in this case, can reduce marginal microleakage and clinical recurrent caries.

Since the association among release of ions such as calcium, fluoride, phosphate, and OH^- with decline in dental caries was not investigated in this study, by considering the obtained results on bioactive restorative materials, clinical assessments of them in oral environment are required.

In conclusion, by considering limitations of this study the mechanical properties of Activa Bioactive and Cention N as new bioactive materials are comparable to composite resins and RMGI, but their storage in an acidic environment can alter ion diffusion behavior and reduce their mechanical properties. Due to the fluoride ion release in acidic environment Fuji II LC is better in clinical conditions, compared to the other materials which were tested in this study.

Disclosure

No potential conflict of interest is relevant to this article.

Author Contribution

All authors actively participated in the discussion of the manuscript's findings, and have revised and approved the final version of the manuscript.

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