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Effects of Incorporation of Marine Derived Hydroxyapatite on the Microhardness, Surface Roughness, and Fluoride Release of Two Glass-Ionomer Cements

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Abstract: Background: The aim of this study was to evaluate the effects of incorporation of hydroxyapatite (HA) derived from cuttlefish bone on the microhardness, surface roughness (SR), and fluoride release (FR) of conventional cure, and resin-modified glass-ionomer cement. Methods: There were four groups for each tested material; experimental glass-ionomer were made by addition and of 2, 5, and 10 wt % HA respectively to conventional glass-ionomers Fuji II LC and Fuji IX GP Extra. One group was prepared without the addition of HA particles. For SR and microhardness measurements sectional Teflon molds (5 mm in diameter and 2 mm deep) were used to prepare 10 samples per group (n = 80). The samples were stored in distilled water at 37 °C for 7 days prior to testing. The SR was measured using a contact type profilometer and the microhardness was determined using a Vickers micro-hardness tester at a load of 980 g for 15 s. For FR measurements, there were six samples per group (n = 48), prepared in Teflon molds (8 mm in diameter and 2 mm deep). The FR was measured with an ionoselective electrode in triplicates after 24 h, 7 days, and 45 days. Statistical analysis was performed using one-way ANOVA with Tukey post-hoc test. Results and Conclusion: Microhardness values obtained for Fuji II modified with 10 wt % HA were significantly higher compared to the other two groups tested. Comparison of materials with respect to SR showed significant difference between them ($p < 0.0001$) with Fuji II and Fuji IX modified with HA having higher SR values. Regarding FR, Fuji IX showed statistically significant higher results than Fuji II, independently of HA modification, and groups modified with 2 and 5 wt % HA showed significantly increased fluoride release in all three time points.

Keywords: glass ionomer cements; hydroxyapatite; microhardness; surface roughness; fluoride release

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1. Introduction

Since their introduction, glass-ionomer cements (GICs) have been a widely used material in restorative dentistry due to their ability to chemically bond to dental tissue, biocompatibility, easy handling, and fluoride release [1–3]. However, disadvantages of GICs—such as poor compressive strength (CS), hardness, elastic modulus, and wear resistance [4,5]—lead to proposing many modifications. One of the attempts is incorporation of different fillers into the matrix of the cement. Recently, the addition of specific percentages of micro- and nano-hydroxyapatite (nano-HA) particles to GICs has shown promising results, such as the increase in adhesive strength to dentin [6] and increased flexural strength, as shown in our previous study [7]. HA derived from fish bone was reported to be biocompatible if incorporated into material [8] and porous spherical HA particles have been shown to increase mechanical properties and the release of fluoride ions most effectively [7,9].

The hardness of restorative material is critical for the clinical longevity of restorations [10]. Previous research has shown that microhardness determined by the Vickers method is a valid measure of the surface properties of GICs [11,12]. An increased roughness can be a predisposing factor for bacterial colonization [13]; when exceeds 0.2 μm, the caries risk is increased because of bacterial accumulation, plaque maturation, and acidity [14]. Therefore, surface roughness (SR) of GIC materials is often defined as a measure of the wear of materials, which strongly influences the success of restoration [15].

The main effects of GIC's anticariogenic and remineralization effects are largely due to fluoride release (FR), which is considered to be an important property of restorative dental materials and its release level has been shown to be influenced by material composition, storage conditions, and curing method [16–18]. Furthermore, it is suggested that addition of nano-sized particles enhances the release of fluoride from the GIC [19].

Having in mind the idea of developing the material with suitable biological and mechanical properties, the aim of this study was to evaluate the effect of incorporation of 2, 5, and 10 wt % HA, derived from the cuttle-fish bone by the hydrothermal method [20] in conventionally cured and light cured GIC, on microhardness and SR of GICs and to investigate its effects on chemical feature and fluoride release. The null hypothesis is that there will be no changes in microhardness, SR and FR properties of GIC after the addition of HA.

2. Materials and Methods

Highly porous HA was obtained from aragonitic cuttlefish (*Sepia officinalis*) bone from the Adriatic Sea by using the hydrothermal method. The HA powder in the form of hexagonal column crystal aggregates with a diameter of <180 μm was prepared by grounding and sifting bones through a 180 μm size sieve [20].

In the present study, two GICs were used: Fuji II LC and Fuji IX GP Extra (GC Corporation, Tokyo, Japan). The HA powder 2, 5, and 10 wt % (three experimental groups) and a glass powder were hand-mixed using a mortar and pestle for 20 min to obtain a homogenous powder. The prepared powder was then mixed with the polyacrylic acid by spatula. Four groups were prepared for each GIC material; the first group was without HA particles, while in the powder of three experimental groups 2, 5, and 10 wt % HA respectively was added.

For microhardness and SR testing sectional Teflon molds (5 mm diameter × 2 mm deep) were used to prepare 10 samples per group (n = 80). After mixing the GIC components by spatulation, the material was poured into a syringe (Centrix, Shelton, CT, USA) and immediately into Teflon molds. To avoid air trapping, polyester strips were placed and the material was gently compressed on both sides of the mold by glass. Fuji IX specimens were left to set for one hour. Both sides of each Fuji II sample were light-cured for 20 s to ensure a proper setting, using a LED lamp (Ivoclar Vivadent AG, Schaan, Lichtenstein, Germany), with intensity 600 mW/cm ^{2}. The specimens were stored in distilled water at 37 °C for 7 days and than tested. SR and microhardness measurements were conducted on the same sample.

SR was measured using a contact type profilometer device (Perthometer M2, Mahr GmbH, Gottingen, Germany). Multidirectional readings were made for each specimen in five different areas. After five sequential measurements were performed at different locations for each specimen, the arithmetic mean of SR was calculated in μm. Specimens were fixed with a special jig to ensure that their position is the same for all measurements.

Microhardness measurements were performed using a digital microhardness tester (HMV-2, Shimadzu Corp., Kyoto, Japan). A 980 N force was applied to the specimens with a diamond indenter for 15 s. The testing machine was calibrated before each measurement. Indentations were made after the specimen surfaces were divided into four quadrants. Two measurements were taken in each quadrant, totaling eight measurements. The mean of the eight measurements represented specimen mean. These points were not at the

margins or areas with visible irregularity. Microhardness was expressed as Vickers hardness number (VHN).

One specimen from each group was sputter-coated with gold and observed under SEM (JSM-6400 SEM, JEOL, Tokyo, Japan) at magnifications X15, X50, and X100.

Samples for the FR measurements were prepared as described for microhardness and SR. Six discs (8 mm diameter × 2 mm thick) for each GIC group were made using a Teflon mold (n = 48). After setting of material, samples were removed from the molds by applying pressure at one side and individually stored in polyethylene vials, in 5 mL deionized water and left at 37 °C for 24 h. The concentrations of fluoride released into the water were measured after 24 h, 7 days, and 45 days in triplicate for each sample and expressed in mg/L (ppm F^-). The deionized water was replaced after every testing procedure. For the measurements, each disk was removed from the water, dried on filter paper, weighed, and immediately immersed in 5 mL fresh deionized water for further measurements. The fluoride concentrations in the water samples were measured using an Ionoselective electrode (F800 DIN, Xylem Analytics Germany, Weilheim, Germany) connected to an ion analyzer (inoLab Multi 9630 DS; Xylem Analytics Germany, Weilheim, Germany). Buffer solution, 0.5 mL of TISAB III (Total Ionic Strength Adjustment Buffer; Merck KGaA, Darmstadt, Germany), was added to the water sample to achieve constant ionic strength and pH.

Regarding a statistical analysis of the data, a descriptive analysis was performed. Normality distribution was checked with the Shapiro–Wilk test while equality of variances was tested with Levene's test. Due to heterogeneity of variances (Levene's test; $p = 0.0265$ for surface roughness and $p < 0.0001$ for microhardness) Welch's one-way ANOVA and a post-hoc Tukey's test were performed. The analysis was conducted using a SPSS statistical package on a Windows platform. The level of significance was set at $p = 0.05$. For FR, obtained data were processed by the method of linear regression analysis, which was recommended by Can Karabulut et al. as the best method for investigation of fluoride releasing materials [21].

3. Results

Descriptive statistics for SR and microhardness properties in the eight groups are shown in Table 1.

Table 1. Descriptive statistics for surface roughness (SR) and microhardness.

Material	N	SR Mean	Microhardness Mean
Fuji II0	10	0.062	0.070
Fuji II2	10	0.069	0.078
Fuji II5	10	0.080	0.087
Fuji II10	10	0.089	0.097
Fuji IX0	10	0.176	0.199
Fuji IX2	10	0.221	0.269
Fuji IX5	10	0.192	0.212
Fuji IX10	10	0.254	0.282

The results of the material comparison are shown in Table 2.

Comparison of materials with respect to SR showed that there is a difference between them ($p < 0.0001$; ANOVA test). The results of the multiple comparison (Tukey test) with respect to SR showed that values for Fuji IX are higher on average in comparison to the values for Fuji II irrespective of HA concentration. For Fuji IX, SR also generally increases with the increase of HA concentration. Roughness of Fuji IX 10 wt % HA was statistically higher than the SR of Fuji IX without HA and Fuji IX 5 wt % HA.

Table 2. Results of the ANOVA test for Fuji II and Fuji IX.

Material	Surface Roughness (Ra)		Microhardness (VHN)	
Fuji II0	0.062	a	55.2	ab
Fuji II2	0.069	a	47.5	bc
Fuji II5	0.080	a	53.0	b
Fuji II10	0.089	a	61.5	a
Fuji IX0	0.176	b	50.6	b
Fuji IX2	0.221	bc	48.9	b
Fuji IX5	0.192	b	47.2	bc
Fuji IX10	0.254	c	40.7	c
p *	<0.0001		<0.0001	

* *p*-value for ANOVA test; a, b, c—materials with the same letter are not significantly different (Tukey test).

There is difference in microhardness between the groups, too ($p < 0.0001$; ANOVA test). The addition of HA to Fuji II is not conclusive. Lowest microhardness is observed for 2 wt % HA, but Tukey test showed that microhardness of Fuji II with 10 wt % HA was significantly higher. In Fuji IX groups, it was observed that increase of HA concentration decreased microhardness. However, only the difference between 10 wt % HA concentration and 2 wt % HA concentration and the difference between 10 wt % HA concentration and group without HA particles added were significant. There was no evidence that microhardness for the group without HA particles and groups with 2 wt % HA and 5 wt % HA differ.

The observed SEM images of representative of one specimen surface from each group are shown in Figure 1.

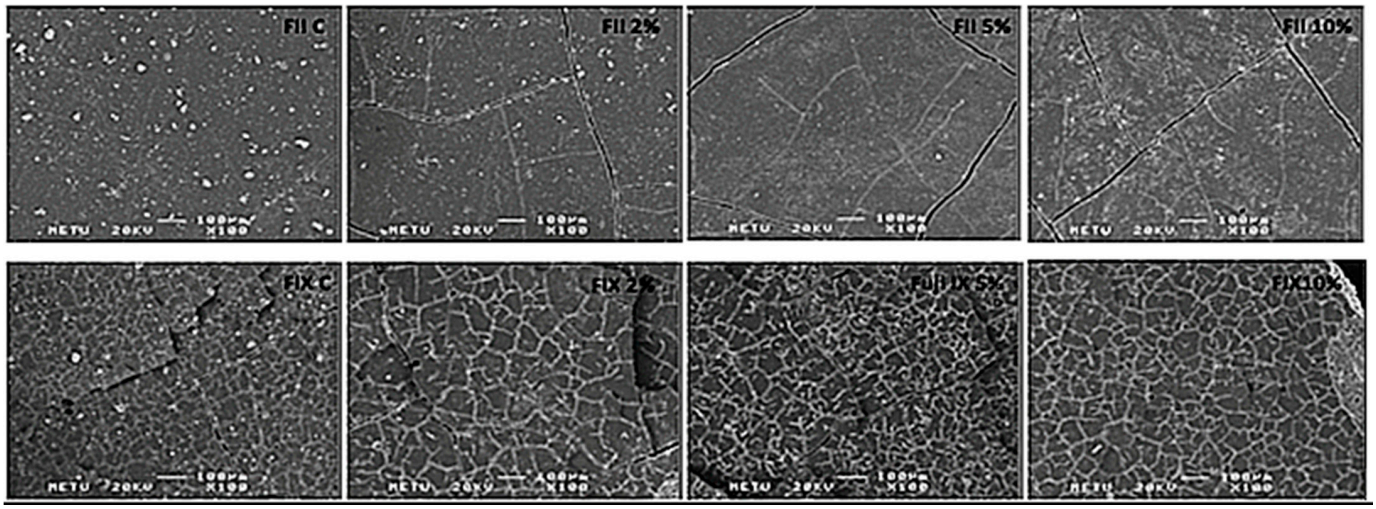

Figure 1. Representative SEM photomicrographs (×100 magnification) of a Fuji II (first row): Fuji II (without HA particles), Fuji II 2 wt % HA, Fuji II 5 wt % HA, Fuji II 10 wt % HA respectively; and Fuji IX (2nd row): Fuji IX (without HA particles), Fuji IX 2 wt % HA, Fuji IX 5 wt % HA, Fuji IX 10 wt % HA respectively.

The SEM images display small glass particles dispersed in the matrix. SEM images of Fuji II, either the group without HA added or experimental groups displayed predominantly smooth, and featureless surfaces (Figure 1, a–d) but cracks or voids were evident on the surfaces of HA added specimens (b–d) in contrast to the group without HA (a). The surfaces of Fuji IX (e–h) appeared relatively rough, demonstrating macro defects on the surfaces.

The results for fluoride ion release are shown in Figure 2.

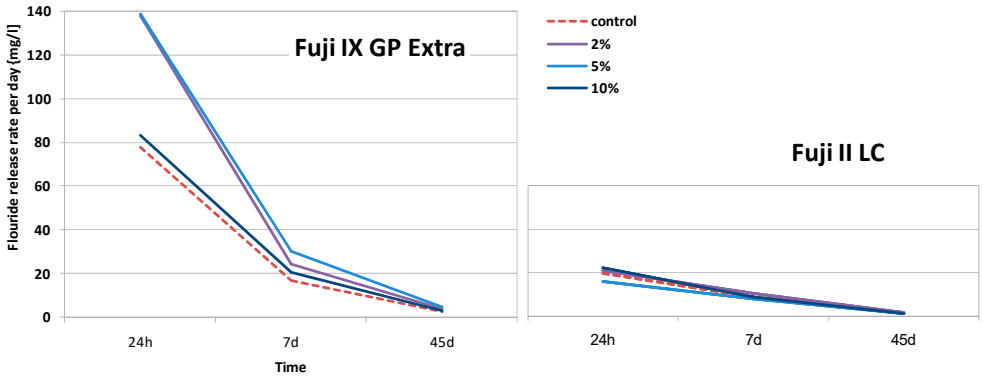

Figure 2. Fluoride release after 24 h, 7 days, and 45 days.

The results for Fuji IX were significantly higher than the results for Fuji II ($p < 0.001$). In terms of time, it can be noticed that the concentration of the released fluoride ions rate significantly decreased with time ($p < 0.001$). This decrease was significantly different for the two materials ($p < 0.001$).

When observing the effects of adding HA to the materials, the fluoride release was different when compared to the group without HA added. This effect was, however, neither consistent nor clearly dependent on concentration. Adding HA in concentrations of 2 or 5 wt % HA resulted in significantly higher FR of Fuji IX in all three time points. The sample with 10 wt % HA showed significantly higher amounts of released fluorides than the group without HA, but lower than the samples with 2 and 5 wt % HA. The results are shown in Table 3.

Table 3. The release rate of flouride after 24 h, 7 days, and 45 days.

Mean ± Standard Error	24 h (mg/L)	7 Days (mg/L)	45 Days (mg/L)	*p*
Fuji IX0	77.7 ± 1.7^{c}	17.1 ± 0.8^{d}	2.7 ± 0.1^{d}	<0.001
Fuji IX2	138.1 ± 1.3^{a}	24.5 ± 0.5^{b}	3.9 ± 0.1^{b}	<0.001
Fuji IX5	138.6 ± 2.3^{a}	30.4 ± 0.2^{a}	4.8 ± 0.0^{a}	<0.001
Fuji IX10	83.3 ± 2.3^{b}	20.8 ± 0.4^{c}	3.3 ± 0.1^{c}	<0.001
Fuji II0	19.6 ± 0.9^{e}	8.9 ± 0.1^{f}	1.4 ± 0.0^{f}	<0.001
Fuji II2	21.1 ± 0.5^{e}	10.4 ± 0.1^{e}	1.6 ± 0.0^{e}	<0.001
Fuji II5	15.8 ± 1.3^{f}	8.1 ± 0.1^{g}	1.3 ± 0.0^{g}	<0.001
Fuji II10	22.1 ± 0.4^{e}	8.8 ± 0.0^{f}	1.4 ± 0.0^{f}	<0.001

$^{a–g}$ the differences between the groups within a specific column/time point are statistically significant with $p < 0.05$.

With Fuji II, the results were similar in the sense that the 2 wt % HA samples showed significantly higher FR rates than the other samples after 7 days and 45 days, while the 5 wt % HA samples had lower results than either 2 wt % HA, 10 wt % HA or samples without HA in all three time points.

4. Discussion

Our results showed that the addition of marine derived HA particles to commercially available GIC materials influenced SR, microhardness and FR, leading to rejection of the null hypothesis. HA particles used for experimental part were micro-sized because it was found that nano-HA-filled materials were rougher [22]. Additionally, micro-particles of HA are easily mixed with resin and nano-HA considerably prolongs the setting time of GICs [23].

SR values for Fuji II were significantly lower in comparison to the values for Fuji IX with no dependence on HA concentration. This is in agreement with the findings of Ismail et al. [24] who showed that resin modified glass ionomer cement (RMGIC) exhibited lower surface roughness than did chemically cured, possibly due to its smaller filler size

(0.02–0.04 μm). SR values increased with the rise of HA concentration in both materials tested as a consequence of HA particles formed by hydrothermal conversion having a cauliflower-like morphology, thus increasing the surface roughness and specific surface energy [20]. Additional SEM analysis confirmed predominantly smooth, and featureless surfaces in Fuji II specimens probably due to incorporation of the resin, while the surfaces of Fuji IX appeared relatively rough demonstrating macro defects on the surfaces faster.

Our results demonstrate a trend of higher microhardness values for Fuji II than for Fuji IX. The exception is value for 2 wt % HA, where mean for Fuji IX sample is higher than mean for Fuji II sample. Yli Up et al. examined the microhardness of the GIC and RMGIC with 10% or 30% HA [25]. They reported that the microhardness of GICs decreased as the amount of HA increased, which is in accordance with results of the present study. The explanation for difference in results could be the fact that the microhardness of HA-added Fuji II increased during water storage [12]. In addition, larger particle size and fewer voids and cracks of RMGIC resulted in higher microhardness values. Among all the eight groups, RMGIC (Fuji II) with 10 wt % HA had the highest microhardness value. When compared to an earlier study where the group with 10 wt % HA had the highest flexural strength value [26], it is possible that this exact concentration of HA particles could improve two important physical properties of Fuji II, microhardness, and SR. This result is in agreement with Lee's survey, which was shown that the physical properties of RMGI improved with the incorporation of 10% nano- and micro-HA [27]. Some studies have shown that incorporation of 5 wt % HA in GICs resulted in improved mechanical properties, including compressive strength and microhardness [28,29]. This difference could be explained with possible non-uniformed glass—HA powder (manual mixing) or inadequate liquid amount, as the volume of HA can change the reaction. It was indeed shown that resin modified GICs are more stable in water than are conventional GICs [28], but resin from RMGICs have been shown to have negative effects on dental pulp, it also exhibits cytotoxicity [30]. It would therefore be ideal in a clinical context to obtain better chemo-mechanical properties of chemically set GICs.

Addition of HA in concentrations of 2 or 5 wt % HA resulted in significantly higher FR of Fuji IX in all three time points

It was previously shown that the light or chemical curing influences fluoride release from resin modified GICs and dual-cured resin cements. Furthermore, photoinitiated polymerization enhances cross-linking density, resulting in the reduced resin matrix permeability for fluoride ions [31], what is in accordance with our results, generally higher FR results in Fuji IX GP Extra than in Fuji II. On the other hand, modification of Fuji II powder with 2 wt % HA positively influenced the fluoride release rates compared to other samples measured after 7 days and 45 days. The modification of Fuji IX GP Extra in all concentrations of HA showed improved FR in all three time points. In our study the highest values of FR were in the first 24 h ranged from 15–138 mg/L for different GICs, what agrees with previous investigations [32,33]. The fluoride ions in the GICs structure do not take part in the setting reaction and after the initial burst effect [34], they are released into the surrounding environment via an ion exchange process. Furthermore, the glass ionomers can also absorb salivary fluoride and act as fluoride reservoirs capable of releasing the ions, which may inhibit caries formation [35].

As HA derived from cuttlefish bone was reported to be biocompatible [16] and a biomaterial without any cytotoxic effect on dental pulp cells if incorporated into restorative material, it could be interesting to perform further investigations about its impact on GICs. The limitations of the study are that it does not implement all the factors that could affect the material itself, such as mastication forces, heat change, exposure to saliva. However, all the samples were exposed to the same experimental conditions. In addition, only representative commercial materials have been used; a wide range of GICs is available and could be tested. Finally, the clinical relevance of our findings remains unclear; these types of in vitro studies provide insight into the possible effects of HA incorporation, but the results should be observed considering the conditions of the study.

5. Conclusions

The present study showed that the addition of micro-HA derived from cuttlefish bone to the powders of Fuji IX and Fuji II does not improve the SR. As far as microhardness is concerned, the addition of HA decreased microhardness values in all groups except the Fuji II 10 wt % HA group. In terms of FR, samples of Fuji IX modified with 10 wt % HA, after 24 h showed most favorable results, and 2 and 5 wt % HA improved FR in all three time points, what can lead to conclusion that the exact concentration of HA incorporated in GIC improving chemo-mechanical properties should be further investigated.

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Informed Consent Statement: Informed consent was obtained from all subjects involved in the study.

Data Availability Statement: The data presented in this study are available on request from the corresponding author.

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