Effect of Hydroxyapatite on Surface Roughness of Zirconomer, and Conventional and Resin-Modified Glass Ionomers

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Article Info	A B S T R A C T
	Objectives: Glass ionomer cements (GICs) are among the most popular dental restorative materials, but their use is limited due to their clinical disadvantages. Many efforts have been made to improve the properties of these materials by adding various fillers. Incorporation of hydroxyapatite (HA) into the GICs is considered to improve the physical properties of restorations, and may prevent treatment failure. This study aimed to evaluate the surface roughness (Ra) of a conventional glass ionomer cement (CGIC), a resin-modified glass ionomer (RMGI) and a Zirconomer with and without micro-hydroxyapatite (μHA).
	Materials and Methods: This experimental study was conducted on 6 groups (n=10) including CGIC, CGIC + μHA, RMGI, RMGI + μHA, Zirconomer, and Zirconomer + μHA. A total of 60 disc-shaped samples (6 mm × 2 mm) were prepared in plastic molds and were stored in distilled water for 24 h. After polishing of the specimens, their Ra was measured by a profilometer in micrometers (μm). The data were analyzed using two and one-way ANOVA, Tukey's HSD test, and independent t-test.
* Corresponding author: Department of Operative Dentistry, School of Dentistry, Shiraz University of Medical Sciences, Shiraz, Iran Email: sbahrani@sums.ac.ir	**Results:** Incorporation of μHA resulted in statistically significant differences in Ra between the study groups (P<0.05). Following the incorporation of μHA, the Ra significantly decreased in CGIC (P=0.013) and Zirconomer (P=0.003). However, addition of μHA to RMGI resulted in a significant increase in its Ra (P<0.001).
	Conclusion: Addition of μHA decreased the Ra of Zirconomer and CGIC, and increased the surface roughness of RMGI samples.
	Keywords: Glass Ionomer Cements; Hydroxyapatites; Surface Properties; Biocompatible Materials

INTRODUCTION

Surface roughness highly affects the erosion and clinical longevity of restorations. A rough surface facilitates plaque accumulation. It also provides a suitable environment for colonization of microorganisms, consequently leading to gingival and periodontal disease and increasing the risk of dental caries [1]. A smooth surface decreases the surface area and surface energy; therefore, accumulation of particles is lower on smooth surfaces [2]. Furthermore, the esthetic success of a restoration is directly correlated with its optical properties. Surface roughness is considered as one of the most important factors affecting the appearance of a tooth-colored restoration [3]. Glass ionomer cements (GICs) are among the popular dental materials

commonly used for direct esthetic restorations in non-load-bearing areas [2]. The original GICs are composed of an aqueous solution of polyacrylic acid, which reacts with a powder consisting of calcium fluoroaluminosilicate glass. These glass fillers are a modification of the former dental silicate cements with an aim to enhance their physical, mechanical and biological properties [4]. The favorable properties of GICs include their optimal biocompatibility, adhesion to the tooth structure, and fluoride release potential. Beside their favorable properties, GICs have some clinical limitations such as long setting time, rough surface structure, and sensitivity to moisture and dehydration during their primary setting [5]. Attempts to overcome these limitations resulted in production of resin-modified glass ionomers (RMGIs) [6,7]. These materials were developed by adding resin monomers to GICs, resulting in much better esthetics, higher mechanical and handling properties, and higher bond strength to enamel and dentin. Another shortcoming of the conventional glass ionomer cements (CGICs) is their low mechanical strength [8]. The main reasons for the application of zirconia as a filler are its good chemical and dimensional stability, high toughness and mechanical strength, and a Young's modulus similar to that of stainless steel alloys. Zirconia has been extensively used for strengthening and toughening of brittle hydroxyapatite, bioglass, and composite materials [9]. A high-strength GIC that is strengthened with zirconia fillers known as Zirconomer, could overcome the disadvantages of the previously used GICs [10]. Hydroxyapatite [$Ca_{10}(PO_4)_6(OH)_2$] (HA) is considered as the main component of the enamel and dentin [11,12]. Owing to its chemical composition and crystallographic structure, it could be used in bone tissue engineering, restoration of periodontal defects, endodontic treatments like pulp capping, apical barrier formation, treatment of primary caries, and also as a filler to reinforce the structure of restorative GICs and composite resins [11,13].

It has been reported that incorporation of HA into GICs can improve some of their mechanical features such as diametral and flexural strength, fracture resistance, compressive strength, and bond strength. It can also remineralize the demineralized dentin and increase its flexural strength [11]. In another study, HA was found to have antibacterial activity against Streptococcus mutans [14]. Due to the favorable biocompatibility of HA and high mechanical properties of Zirconomer, incorporation of HA as filler into Zirconomer has gained interest [9]. A recent study by sharafedin et al. [15] showed that addition of 5 and 15wt% micro-hydroxyapatite (μHA) to Zirconomer and RMGI increased their surface microhardness. Although the surface roughness of GICs and the factors affecting it have been previously evaluated in many studies [16–20], and efforts have been made to improve this property of GICs [2], limited studies have evaluated the effect of incorporation of μHA on the surface roughness of GICs. Thus, this study was designed to investigate the effect of incorporation of μHA on the surface roughness of three GICs namely CGIC, RMGI, and Zirconomer.

MATERIALS AND METHODS

The study protocol was approved by the Ethics Committee for Research of Shiraz University of Medical Sciences (IR.SUMS.REC.1394.S360). In this experimental study, 60 samples were fabricated in six groups (n=10). Disc-shaped samples were made by using cylindrical plastic molds measuring 6 mm × 2 mm. Table 1 presents the characteristics of the materials used in this study. Sample preparation in the six groups was as follows:

Group 1 (CGIC): To prepare the CGIC samples based on the manufacturer's instructions, one scoop of CGIC powder and one drop of the liquid (Fuji II GC, GC Corp., Tokyo, Japan) were mixed on a cold clean glass slab using a plastic spatula for 25 s. The mixture was applied into the mold; the upper and lower surfaces were covered with transparent Mylar strips (Fintrec Transparent Matrix, Pulpdent Corp., Watertown, MA, USA), and were then placed between two glass slabs for complete setting. The material set within 5.5 min after mixing. A thin layer of copal varnish (Kimia, Tehran, Iran) was applied over the surface.

Table 1. Composition of materials used in this study

Materials (batch number)	Manufacturer	Composition
Fuji II GC (1407141)	GC Corp., Tokyo, Japan	*Powder:* fluoroaluminosilicate glass *Liquid:* polyacrylic acid, itaconic acid, tartaric acid, maleic acid, water
Fuji II LC (1407021)	GC Corp., Tokyo, Japan	*Powder:* aluminofluorosilicate glass, urethane dimethacrylate, camphorquinone *Liquid:* polymer acrylic acid, distill water, 2-hydroxyethylmethacrylate
Zirconomer (04140781)	Shofu INC, Kyoto, Japan	*Powder:* aluminofluorosilicate glass, zirconium oxide, tartaric acid *Liquid:* polyacrylic acid, deionized water
Hydroxyapatite (5465141Q788)	Sigma-Aldrich, USA	Calcium hydroxyphosphate
Varnish (01071020)	Kimia, Iran	Copal resin, ethanol

Group 2 (CGIC + µHA): Each sample in this group contained 85wt% CGIC powder and 15wt% µHA (Sigma-Aldrich, St. Louis, USA), which were separately weighed by using a scale with 0.0001 g accuracy (GR+360; A&D, Tokyo, Japan).

Fig. 1. Study groups based on the materials

The powders were blended on a glass slab using a plastic spatula. The mixture was transferred to clean amalgam capsules and mixed again in an amalgamator (FD-4300; Faghihi, Tehran, Iran) for 20 s to obtain uniform and equally homogenized powder in all samples. The obtained powder was mixed with CGIC liquid similar to group 1. The mixture was then applied into the molds and allowed to set for 5.5 min, and was finally coated with varnish.

Group 3 (RMGI): The samples in this group were made of one scoop of RMGI (Fuji II LC, GC Corp., Tokyo, Japan) and 2 drops of liquid that were mixed for 25 s according to the manufacturer's instructions. The mixture was inserted into the molds and covered with Mylar strips as in group 1. It was placed between two glass slabs, and cured with a LED curing unit (Bluelex GT 1200; Monitex, San-Chong, Taiwan) with 1200 mW/cm² light intensity for 20 s. The tip of the device was in contact with the slab to achieve better cure. Finally, a thin layer of varnish was applied over the cured samples.

Group 4 (RMGI + µHA): The powder containing 85wt% RMGI and 15wt% µHA was prepared as in group 2.

Group 5 (Zirconomer): The same as group 1, two scoops of Zirconomer (Zirconomer, Shofu Inc., Kyoto, Japan) powder were mixed with one drop of liquid for 30 s. The mixture set within 3.5 min, and coated with varnish.

Group 6 (Zirconomer + µHA): As in group 2, the powder containing 85wt% Zirconomer and 15wt% µHA was mixed with Zirconomer liquid. Figure 1 displays the study groups based on the materials.

All the fabricated samples were removed from the molds and stored in distilled water at room temperature for 24 h.

Then, the surface of the samples was polished using multistep polishing paper discs (Super Snap Rainbow Technique kit; Shofu Corp., San Marcos, USA) and low-speed handpiece under water coolant. Each specimen was polished for 30 s at each step. Polishing was performed with a rotational planar motion around the normal vector of the surface [21] To eliminate the debris, the samples were rinsed with distilled water for 1 min in an ultrasonic bath (Renfert GmbH, Hilzingen, Germany).

The surface roughness (Ra) was measured at three points of each sample, using a surface roughness tester (Rugosurf 20; Tesa Tec, Renens, Switzerland). The mean of the three values was recorded as the final surface roughness of each sample. The mean value of each group was used for statistical analysis. The study groups were analyzed by two and one-way ANOVA, Tukey's HSD test, and independent t-test using SPSS version 24 (SPSS Inc., Chicago, IL, USA) (P<0.05).

RESULTS

Two-way ANOVA revealed significant interaction effect of the type of GIC and μHA application on Ra (P<0.001). As shown in Figure 2, addition of μHA had different effects on the mean Ra values in different types of GICs. Therefore, subgroup analyses were performed to compare different groups.
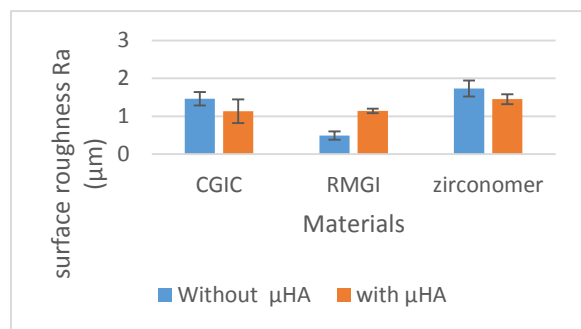

Fig. 2. Mean and standard deviations of the surface roughness of materials with and without μHA. two-way ANOVA revealed a significant interaction effect between the study groups

One-way ANOVA showed significant differences in Ra between the groups (P<0.001).

The Tukey's HSD test revealed that the mean Ra value in the RMGI+μHA group was slightly higher than that in the CGIC+μHA group; however, this difference was not statistically significant (P=0.992). The Ra in Zirconomer+μHA group was significantly higher than that in RMGI+μHA (P=0.016) and insignificantly higher than that in CGIC+μHA (P=0.12) groups. Without adding μHA, the mean Ra value of Zirconomer was significantly higher than that of RMGI (P<0.001) and CGIC (P=0.05). Furthermore, the mean Ra value of CGIC was significantly higher than that of RMGI without μHA (P<0.001). Independent t-test showed that while addition of μHA to the testing materials resulted in a significant decrease in the Ra values of Zirconomer (P=0.003) and CGIC (P=0.013), a significant increase was observed in RMGI following μHA addition (P<0.001). It should be noted that the lowest and the highest Ra values were recorded for RMGI (0.49 μm) and Zirconomer (1.73 μm), respectively. Table 2 summarizes the mean±standard deviation of Ra in each group.

Table 2. Mean and standard deviation (SD) of surface roughness values (Ra in μm) of the study groups

Groups	Mean±SD		P*
	- μHA	+ μHA	
CGI	1.46±0.18[A]	1.13±0.31[A]	0.013
RMGI	0.49±0.11[B]	1.14±0.06[A]	<0.001
Zirconomer	1.73±0.21[C]	1.45±0.13[B]	0.003
P**	<0.001	0.006	---

In each column, the mean Ra values with different uppercase letters are significantly different (Tukey's HSD test). *P: independent t-test, **P: one-way ANOVA F test CGIC: conventional glass ionomer cement; RMGI: resin-modified glass ionomer, μHA: micro-hydroxyapatite

DISCUSSION

It has been reported that incorporation of HA could improve some physical characteristics of GICs [15].

Surface roughness is considered as one of the most important parameters of dental restorations. In the present study, we investigated the effect of adding HA on surface

roughness of GICs. Micro-HA was chosen in this study because it reinforces the GICs and could easily mix with resin (either Bis-GMA+HEMA or Bis-GMA+TEGDEMA). Although nano-HA has a more similar crystal size to the mineral phase of the tooth structure, nano-HA considerably increases the setting time of GICs [15]. It has also been reported that the surface mechanical properties of nano-GICs are not superior to those of GICs without nanofillers [22].

Surface roughness can be evaluated through different methods such as visual evaluation under an optical microscope, scanning electron microscopy, atomic force microscopy, and profilometric analysis [16]. The current study benefited from profilometric analysis to measure Ra due to the optimal accuracy, applicability, and simplicity of this method. Ra is the algebraic average height of the roughness component irregularities from the mean line measured within the sampling length. It should be noted that higher Ra values indicate higher surface roughness [2] Three profilometric measurements were made on each sample, and the mean value was recorded as the Ra of the respective sample in this study. The current study measured the mean Ra values to be 0.49-1.73 µm for specimens made of GICs without µHA and 1.13-1.45 µm for GICs mixed with µHA. Apparently, the material composition caused changes in the Ra values. Among all the tested GICs, RMGI had the lowest Ra value, which was in accord with other studies [2,18,23].

It has been reported that materials with larger particle size have a rougher surface [24,25]. Thus, particles size is a determining factor in the surface roughness of GICs [26]. The particle size of RMGI is smaller than that of Zirconomer [27] and CGIC [25]; therefore, it can justify the lower Ra of RMGI compared with Zirconomer and CGIC. In addition, presence of resin component contributes to a reduction in RMGI surface roughness. Resin improves the surface properties of GICs; thus, in presence of resin in GICs, smaller particles are detached from the material during the polishing process, and a smoother surface is achieved [18]; therefore, it probably causes

smoother surface of RMGI after polishing.

According to the results, Zirconomer had the highest Ra value compared with other groups (regardless of the presence of µHA), which was in agreement with previous studies [28,29]. Higher Ra values in Zirconomer may be attributed to its larger particle size [25]. The mean Ra value in the CGIC group was between those of Zirconomer and RMGI samples. In line with the study by Gladys et al, [25] the mean particle size in CGIC was higher than that in RMGI and lower than that in Zirconomer.

The interaction between GIC and HA occurs through Ca ions in HA and the carboxylate group in acrylate polymer. The bond between GIC and HA is also affected by the adsorption between carboxylic acid and HA, as well as the interaction of the functional groups of polymer and HA. Due to their contribution to the reaction between the powder and liquid in GIC, HA particles probably participate in acid-base reactions through ion release [11]. Seemingly, in the present study, chemical curing increased the setting time of Zirconomer (3.5 min) and CGIC (5.5 min) compared with RMGI, which was light-cured. In other words, HA particles and glass-ionomer have more time to interact in CGIC and Zirconomer groups, probably resulting in a more homogenized mixture than RMGI + µHA leading to lower surface roughness. However, based on some reports, HA particles are likely to act as a barrier, preventing the bonding of components in light-cure RMGI [11,30], which can be the cause of increased Ra in µHA-containing RMGI.

The ability to interact with HA led to smoother surface of HA-incorporated CGIC and Zirconomer compared with groups devoid of HA, which could be due to the small particle size of µHA. Polishing, its technique, and polishing tools are among other factors affecting the surface roughness. Although use of a Mylar strip results in the smoothest surface possible compared with any finishing treatment, some degrees of finishing and polishing are often required clinically [19]. Previous studies indicated that multi-step polishing would yield the best results [20,31],

and also lower Ra values could be obtained in polishing with planar motions for any type of material composition and abrasive disc [21]. It has also been reported that polishing with aluminum-oxide discs results in a polished surface without dislodging of glass particles [31,32].

In the present study, according to the clinical conditions, multiple polishing instruments were used, and polishing was carried out with aluminum-oxide discs in a planar motion to achieve a better polished surface. Each specimen was polished for 120 s in four consecutive steps. Polishing was accomplished by four flexible discs with different grits to make uniform abrasion in the matrix and filler of samples. GIC with incorporated μHA is suggested as a potential dental restorative material. More investigations are recommended on the effects of HA with different amounts and particle sizes on surface roughness by simulating the oral conditions.

CONCLUSION

Considering the limitations of the present study, it can be concluded that addition of μHA had a significant effect on the surface roughness of CGIC, RMGI, and Zirconomer. Hence, the material composition can be a determining factor in the Ra value. It was found that incorporation of μHA may reduce the surface roughness of Zirconomer and CGIC, but it can increase the Ra of RMGI. RMGI yielded the smoothest surface in comparison with other groups. Moreover, Zirconomer showed the highest surface roughness among the tested materials.

ACKNOWLEDGMENTS

CONFLICT OF INTEREST STATEMENT

None declared.

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