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Bioactivity, Water Sorption and Solubility of Sol-Gel Formulated Glass-Ionomer Cement Modified With Nano-Fluorapatite

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Abstract

OBJECTIVE:

To assess the bioactivity, water sorption and solubility of sol-gel formulated glass-ionomer cement (FGIC) (60%SiO_2, 35%CaO, 5%P_2O_5) with and without incorporation of Nano-fluorapatite (Nano-FAp) regarding to the commercial glass ionomer cement (CGIC).

MATERIAL AND METHODS:

The tested glass-ionomer cement was formulated by sol-gel method and nano-fluorapatite was incorporated into this formulated glass-ionomer with different percentages (1wt%, 2wt%, 3wt%, and 4wt %). A total number of 216 samples were used in this study. The samples were categorized in six groups; group1 CGIC (negative control), group 2; FGIC (positive control), group 3; FGIC/1wt% Nano-FAp, group 4; FGIC/2wt% Nano-FAp, group 5; FGIC/3wt% Nano-FAp; and Group 6; FGIC/4wt% Nano-FAp. Each group was subdivided into two subgroups of (18) samples each according to type of the test.The samples were characterized and tested for bioactivity after immersion in simulating body fluid (SBF) for 28 days, and for water sorption and solubility after 7 days of immersion in distilled water. One-way ANOVA tests was used to analyze data statistical significant is stablished at probability level of 0.05.

RESULTS:

FGIC recorded higher Ca/P ratio when compared to CGIC and this ratio increases with the increase of the percentage of the added nano-FAp. Also, FGIC recorded higher water sorption and solubility when compared to CGIC and its water sorption and solubility decreased when the amount of added nano-FAp increased.

CONCLUSION:

The sol-gel formulated GIC has higher bioactivity, higher water sorption and solubility than the commercial GIC. However, the incorporation of nano-FAp improves the bioactivity of this sol-gel formulated GIC and decreases its water sorption and solubility along with the increases in concentrations.

I. Introduction

Commercial glass ionomer cement (GIC) usually used as a restorative and lining material for both primary and permanent teeth due to its ability to release fluoride, biocompatibility, close matching thermal coefficient, and adhere chemically to the hard tooth structure . However, it has some limitations such as lower mechanical properties, high solubility and lower bioactivity [1, 2].

The sol-gel synthesis method for glass considered as a useful alternative to the traditional melt and quench method through using a relatively low temperature method of the oxide elements which usually below the crystallization temperature via utilizing liquid chemical precursors to produce glasses and/or ceramic with high homogeneity and purity [3].

Recently, marked improvements were carried out in order to enhance the physical properties and bioactivity of this traditional cement through addition of other nano-ceramic materials such as nano-fhuoroapatite (FAp) and nano-hydroxyapatite (HAp) into the conventional GIC [4].

However, fluorapatite has chemical structure similar to hydroxyapatite, but it has higher mechanical strength, greater thermal stability, and lower solubility in acids than hydroxyapatite [5, 6]. Fluorapatite categorized as a biomedical material owing to its favorable chemical durability, low solubility, and excellent biocompatibility [7].

Furthermore, it was suggested that addition of nano-fluorapatite with sufficient amount into the GIC which prepared via sol-gel method could improve its physico-mechanical properties as well as its bioactivity [3, 8].

Therefore, the present study was conducted to evaluate the bioactivity, water sorption and solubility of sol-gel formulated glass ionomer cement and the effect of incorporation of different nano- FAp concentrations on these properties.

II. Material and methods:

The materials used in this study were, commercial glass ionomer cement powder and liquid (GC gold label, Tokyo, Japan), Sol-gel formulated glass ionomer cement (60% SiO_2, 25%CaO , 5% P_2O_5) and nano-floroapatite (Egyptian Nano Gate Company).

Grouping of samples:

A total number of 216 samples were used in this study. The samples were divided into six groups according to modifications:

Group1: CGIC powder mixed with commercial GIC liquid.
Group 2: FGIC powder mixed with commercial GIC liquid.
Group 3: FGIC powder 1wt% nano-FAp powder mixed with commercial GIC liquid.
Group 4: FGIC/2wt% nano-FAp powder mixed with commercial GIC liquid.
Group 5: FGIC/3wt% nano-FAp powder mixed with commercial GIC liquid.
Group 6: FGIC/4wt% nano-FAp powder mixed with commercial GIC liquid.

Each group was subdivided into two subgroups of (18) samples each according to type of the test (bioactivity and water sorption & solubility).

2.1. Preparation of glass powder by sol-gel method:

For preparation of 100gm of sol gel FGIC powder (60%SiO_2, 25%CaO and5% P_2O_5) various reagents and solutions with specific concentration were selected as a source supply of SiO_2, CaO and P_2O_5 (all chemicals used were manufactured by Sigma-Aldrich, St. Louis, MO, USA). The solutions were mixed together in electrical magnetic stirrer with continuous stirring in the following order: 26.134ml Tetra Ethyl Ortho Silicate (TEOS), 23.384ml distilled water, and 4.19ml nitric acid (HNO_3) together in ethanol at 1500.rpm for 45 minutes, then 1.204ml Tetra Ethyl Phosphate (TEP) and 8.507gm calcium nitrate tetra hydrate {$Ca\ (NO_3)_2 \cdot 4H_2O$} were added to mix and allowed to react for further 45 minutes. After that, all reagents were left under continuous magnetic stirring for 60 minutes to complete hydrolysis. Finally, 4.19ml ammonia solution (as gelation catalyst) was dropped into the mixture and manually flipped with glass rod to prevent the formation of bulked gel [3].

Thereafter, the prepared gel was left to dry at 100° C-120° C for 2 days and then sintered at 580° C for 2 hours in electrical oven. Then, the sintered powder was ground into fine powder using laboratory mortar and pestle and then was passed through a sieve with a mesh number 325 mesh to give particle size less than 45 um [3].

2.2. Preparation of modified sol-gel glass powder:

Nano-FAp (Nano Gate Company, Egypt) were added to the previously prepared glass powder with concentrations of 1 wt. %, 2 wt. %, 3 wt. % and 4 wt. % via using a sensitive digital balance accurate to 0.0001 mg (KERN ABJ 220-4NM, KERN & SOHN GmbH, Balingen – Germany) and then mechanically mixed with dental amalgamator for 30 s, separately to obtain a homogenous distribution of the incorporated nanoparticles [8].

2.3. Characterization of unmodified and modified sol-gel glass powder:

Characterization of the unmodified and modified powders was made by X-ray diffraction (XRD) {Siemens D5000} to identify the nature of glass (amorphous or crystalline). Also, Fourier transform infrared (FTIR) spectroscopic analysis **(Thermo scientific™ Nicolet™ iS™50 FTIR)** was used to determine the functional group.

2.4. Sample preparation:

The samples of etch group were prepared using special designed Teflon molds with diameter and thickness of 7 mm X 1.5 mm for bioactivity test and 9mm X 2 mm for water sorption and solubility test [9,10]. The powder of each group was mixed with commercial glass ionomer liquid according to the manufacturer instructions. Then, each mixed cement was placed in the Teflon mold on glass plate then covered with celluloid strip, pressed with another glass plate and left for setting.

2.5. Bioactivity test:

The samples were immersed in Hank's balanced salt solution as SBF and stored in an incubator at 37°C for 28 days (the solution was changing every 3 days to keep ion concentration at the same level). At the end of storage time, all samples were collected and imaged by scanning electron microscope (SEM) and analyzed for apatite formation (Ca/P ratio) by Energy Dispersive X-Ray (EDX) (Photographic image JEOL JSM-6510LV microscope). The results of this test were presented and descried as numerical values [9].

2.6. Water sorption and solubility test:

The set samples of each group were weighted in an electronic digital balance with microgram readability to record the initial weight (ml), then samples were stored in tubes with distilled water and held in stand at 37 ± 1°C for 1 week in an incubator (Fisher scientific ,200 Series, Model 255D, U.S.A) and the wet weight was recorded (m2). Then, the samples were then dehydrated using desiccator at 37 ± 1°C for 24 h and weighed again (m3). The water sorption percentage was gained from the difference between the recorded initial weighing and the recorded wet weighing (m2-m1) by the following equation: Sorption (%) =m2-m1/m1 x100. While, the solubility percentage (material loss) was calculated from the difference between the initial and final drying weight of each sample (m1-m3) by the following equation: Solubility (%) = m1-m3/m1 x100 [10].

2.7. Statistical analysis:

PASW Statistics 17 (SPSS Inc., Chicago, IL, USA) soft wear was used for all statistical tests with level of significance level at 0.05. ANOVA test were used to compare the means of the recoded values followed by pair wise comparison of means between the tested groups.

III. Results:

3.1. Characterization of the sol-gel prepared cement:

3.1.1. XRD analysis:

The XRD patterns of prepared glass powders (a) exhibit no diffraction peaks were observed indicating amorphous structure of prepared glass powder. While, the XRD pattern of the prepared glass powders with nano-FAp (b, c, d, e) showed markedly broader peak at $2\theta = 33^0$ with slight shift of this peak to the right-hand side in the fluoridated samples which indicates the substitution of OH with F ions in the apatite structure. This shift has been related to the a-axis contraction caused by smaller size of F ions compared to OH ions (Figure 1).

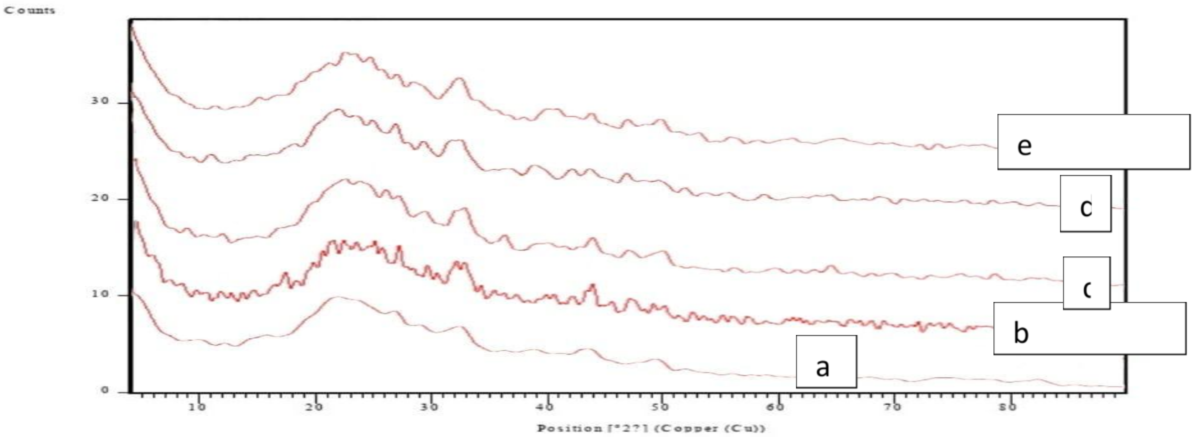


Figure (1): XRD patterns of different prepared glass powders.

3.1.2. FTIR analysis:

The peak located at 467 cm-1 is characteristic to bending vibration mode of Si – O, also peaks at 875 cm-1 and 1099 cm-1 corresponding to symmetric stretching vibration of Si – O, respectively, asymmetric stretching vibration of Si – O – Si bridges. The presence of phosphate in glass structure was evidenced by the peaks located at 565 cm-1 encountered in case of P – O bending vibrations of $PO_4{}^{3-}$ structure. The high $Ca_2{}^{+}$ content also causes the formation of a band at 1432 cm-1.The broaded band located at 3441 cm-1 corresponds to the stretching vibration of hydroxyl (O - H) common especially in case of silanol groups (Si – OH) and chemically bound water presents in the SiO_2 – Ca_O – P_2O_5 glass structure. The presence of H_2O molecules in the same glass structure has been confirmed by the peak at 1638 cm-1. Peaks located at 1384 cm-1 are characteristic to nitrate groups present in the structure of glass, through the use of Ca $(NO_3)2 \cdot 4H_2O$ as a precursor in the synthesis of this material On the other hand, the small peaks at 2426, and 2923 cm^{-1} spectrum are related to presence of C-H (υ) group and were weakly observed. Moreover, the analyzed composition powder had a broad and well-defined peak at 3441 cm^{-1} which represent Si-OH group was observed. On the other hand, the nan-FAp modified groups revealed that the small peak at 2426 cm^{-1} spectrum is related to presence of C-H (υ) group and was absent in 3% and 4% Nan-FAp modified groups (Figure 2).

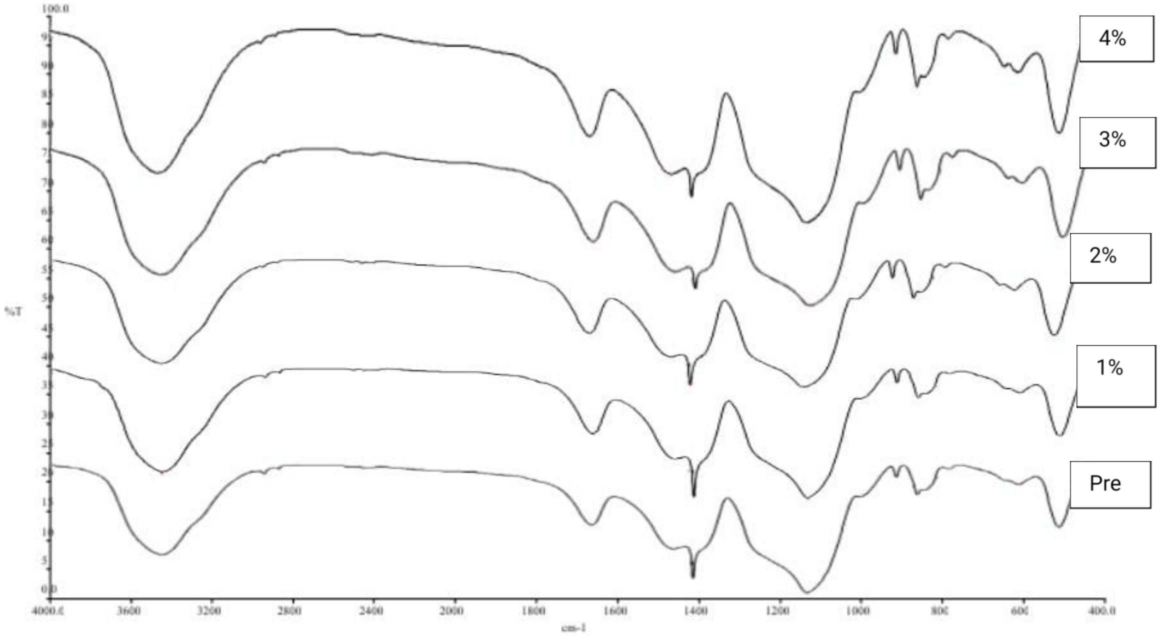


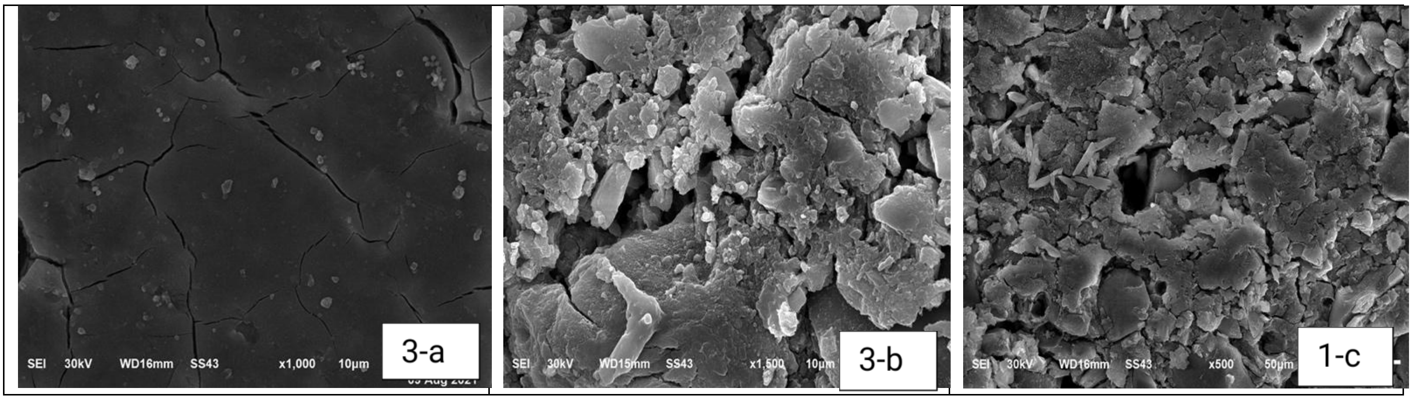
Figure (2): FTIR spectra of different prepared glass powders.

3.2. Bioactivity results:

3.2.1. SEM and EDX Examination:

After immersion in SBF for 28 days the SEM image of the commercial GIC samples (group 1) showed discrete small apatite crystals formed on the cement surface with Ca/P is 1.60, which is lower to stoichiometric value of hydroxyapatite (1.67) (Figure 3-a).The SEM image of the sol-gel prepared (group 2) revealed contiguous spreading of nuclei layer formed on the surface of the prepared glass cement samples with ratio of Ca/P is (1.65) (Figure 3-b).

However, the images of FGIC/1% Nano-FAp (group 3) represent more homogenous and continued network of nuclei layer covered the surface of the prepared glass cement with ratio of Ca/P is (1.66), which is relatively near stoichiometric value (Figure 1-c). Also, the images of FGIC/2% Nano-FAp (group 4) exhibited continued network of submicron nuclei layer that interconnected and cover the cement surface with ratio of Ca/P is (1.83), which is higher than stoichiometric value (Figure 3-d). Moreover, the images of FGIC/3% Nano-FAp (group 5) showed more homogenous and continued network of nuclei layer that hides the cement surface with ratio of Ca/P is (1.95) (Figure 3-e). Furthermore, the images of FGIC/4% Nano-FAp (group 6) revealed more homogenous and continued network of apatite layer with honey-comb like appearance and considerable thickness covered the cement surface with ratio of Ca/P is (2.11) (Figure 3-f).



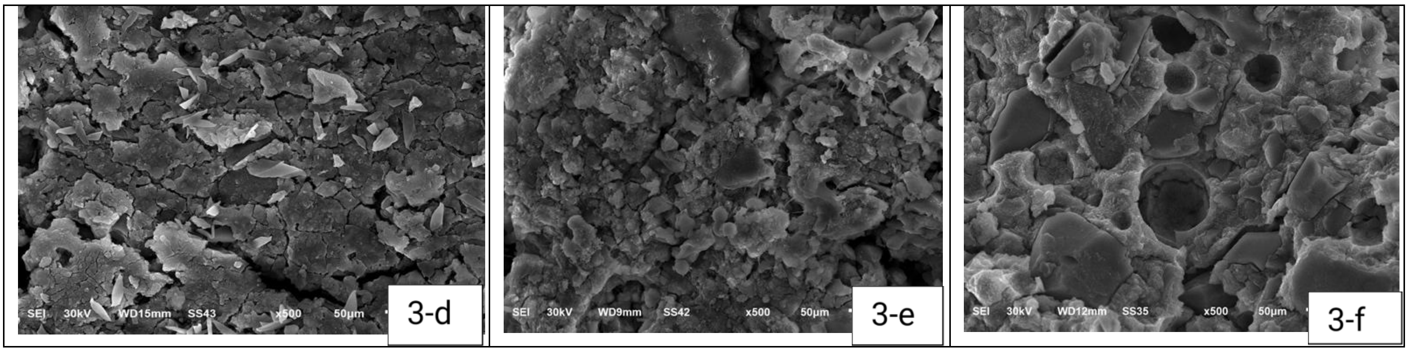


Figure (3): SE Micrographs of the CGIC (a), FGIC (b), FGIC/1% Nano-Fap (c), FGIC/2% Nano-FAp (d), FGIC/3% Nano-FAp (e), and FGIC/4% Nano-FAp (f) after immersion in SBF solution for 28 days.

3.3. Water sorption results:

The mean values and standard deviation of water sorption percentages for all groups are summarized in table (1).

One way ANOVA test showed that there was astatically insignificant difference among mean values of all groups. CGIC (group 1) should lower water sorption percentage when compared to GIC (group 2) and incorporation of nano-floroapatite insignificantly reduce water sorption percentage in the remaining groups.

Table (1): Comparison of water sorption (%) of all tested groups.

Sorption	Group 1	Group 2	Group 3	Group 4	Group 5	Group 6
Mean ± SD	1.82 ± 2.00	5.96 ± 0.67	5.36 ± 0.46	4.66 ± 0.56	4.31 ± 5.41	3.61 ± 2.85
F test	1.506					
P value	0.225 ^{ns}					
P1		0.051^{ns}	0.062^{ns}	0.104^{ns}	0.151^{ns}	0.296^{ns}
P2			0.728^{ns}	0.449^{ns}	0.338^{ns}	0.176^{ns}
P3				0.680^{ns}	0.538^{ns}	0.308^{ns}
P4					0.838^{ns}	0.539^{ns}
P5						0.681^{ns}

; ns = non-significant.

3.4. Water solubility results:

The mean values and standard deviation of water solubility percentages for all groups are summarized in table (2).

One way ANOVA test showed that there was statically significant difference among mean values of all groups. CGIC (group 1) should lower water solubility percentage when compared to GIC (group 2) and incorporation of nanofloroapatite significantly reduce water solubility percentage in the remining groups.

Table (2): Comparison of water solubility (%) of all tested groups.

Solubility	Group 1	Group 2	Group 3	Group 4	Group 5	Group 6
Mean ± SD	5.80 ± 0.85	9.58 ± 1.58	9.25 ± 0.45	8.86 ± 0.51	8.12 ± 0.31	7.09 ± 1.39
F test	10.883					
P value	0.001*					
P1		0.001*	0.001*	0.001*	0.001*	0.049*
P2			0.601	0.254	0.027*	0.001*
P3				0.529	0.080	0.002*
P4					0.246	0.009*
P5						0.107

; The result is significant at $p < 0.05$.
; The result is non-significant at $p > 0.05$.

IV. Discussion:

The sol-gel synthesis method was selected in the present study as route for production of the glass compositions because the sol-gel method serves as a useful alternative to the conventional melt-quench method and refers to a low- temperature synthesis method (usually below the crystallization temperature of the oxide elements) by using chemical precursors in liquid form to produce ceramics and glasses with high purity and homogeneity [3]. However, a comparison between sol-gel and melt-derived glasses with similar compositions also appear to have similar structure and atomic correlations [11].

Also, this sol-gel technology used for the development of the glass which prepared in this study depends mainly on precursor compounds such as tetra ethyl ortho silicate and calcium nitrate tetra hydrate that have high solubility in an organic solvent that can easily transform into chemically reactive forms are essential [12]. Moreover, in this study the glass was formulated as quaternary phosphate-based glasses system with high silica content were successfully prepared by using P_2O_5 as a phosphorus precursor which dissolved in anhydrous ethanol [11, 13].

Furthermore, Nano-FAp were added to the newly synthesized glass to modify its physical and biological properties as it was reported that the addition of highly crystalline particle into GIC exhibited superior mechanical properties and bioactivity than the original GICs [4,5,14]. However, it is necessary to control the fluoride content in the apatite lattice to achieve the best biological and physical performance [6, 15]. As it was found that the increase of fluoride content could adversely affect the physical and the biological performance of the modified glass [5, 6, 15, 16]. Therefore, in the present study the Nano-FAp were added to the prepared glass with percentage between (1-4%) to determine the proper concentration of the Nano-FAp that improve the performance of the prepared glass without any adverse effect.

The potential of apatite formation on the cements surfaces in the presence of SBF was considered as a measure of their bioactivity behavior [6]. Therefore, in the present study the bioactivity of the commercial GIC and the newly synthesized glass were compared regarding to their bioactivity via immersion for SBF for 28 days.

In the present study the XRD analysis results of the newly synthesized glass in the present study showed that the prepared glass was amorphous structure. This agreed with some investigators, [17] as they reported that the sol-gel preparation process used usually to obtain a homogeneous and amorphous gel solid from a liquid sol. The glass structure of the synthesized GIC powder was confirmed by the results of FTIR analysis by the presence of well-defined bands at 467 cm^{-}1 and at 875 cm^{-}1 which is characteristic for Si-O-Si and Si-O that represent the main constitutional composition of GIC. While, the spectral band at 1432 cm^{-1} represent the Ca group [18].

Moreover, the diffraction peaks are markedly broader suggesting that the prepared powder particles were in Nano-size [19]. The improved crystallinity of FA may come from the driving force provided by the high affinity of F- groups for the apatite crystal growth during precipitation [19, 20]. Additionally, the FTIR analysis of the modified glass groups indicates the presence of well-defined bands at 565 vibration modes for the PO4^{3-} group indicates the apatite structure of the incorporated fluorapatite. Also, a broad well-defined peak at 3441 cm^{-1} related to the OH group of the apatite in fluorapatite was clearly observed [18, 21].

According to the results of the present study the conventional GIC has lower ability to form apatite layer when compared to the newly prepared glass cement. Presumably this due to the adsorption of PO4^{3-} by hydrogen bonding and Ca^{2+} ions on the silica surface induce the precipitation of the apatite-like layer [22]. This may be because of for conventional GIC, it is considered difficult to form such apatite layer because the release of PAA from the GIC lowers the pH and inhibits the formation of apatite [23].

However, the high bioactivity of the sol–gel glass is mainly due to the highly porous structure of the glass, which is somewhat different from the case in the melt- derived glass [24]. Some investigators, [25] reported that the rapid cation exchange of Na^{+} and/or Ca^{2+} with H^{+} ions as well as phosphate (PO4^{3-}) between the sol-gel glass and SBF solution due to surface hydrolysis. Then, the solution acidity increases gradually, and a silica-rich region forms on the surface of glass cement [26, 27]. Soluble silica is lost in the form of Si (OH) 4 to the solution and repolymerizes in a silica-rich layer [25].

However, the results of the present study showed that the incorporation of fluorapatite in the newly prepared glass cement resulted in increase in its bioactivity respectively with the increase of the concentration of the incorporated nanoparticles. This due to the presence of fluorine ion in the fluorapatite structure that has lower solubility [28]. Positive cations such as calcium as well as positive hydrogen were replaced in the environment. In addition, phosphate and fluorine ions replaced the hydroxyls, and the ion exchange inside the solution changed significantly [26, 28].

The increased water sorption of the newly synthesized glass in this study may be because of presence of pores in the synthesized glass due to gelation and water evaporation [29]. As the free volumetric theory could explain this water sorption among the existed porosity, whereby the water diffuses through micro-voids without any mutual relationship with the polar molecules in the material [30].

However, the increased solubility of the newly formulated glass may be due to its composition. As the synthesized phosphate-based glasses have the potential to dissolve completely in an aqueous medium due to phosphate content which increases its solubility [31, 32]. However, the increasing Ca and silicate content in the commercial GIC decreases its solubility when compared to the sol-gel prepared glass which have phosphate content [30, 33].

However, the results of the present study showed that the solubility of the newly synthesized glass was decreased respectively with the increase of the amount of the incorporated fluorapatite. This might be due to stability and slow solubility of fluorapatite in distilled water [34]. Also, the low solubility of fluorapatite modified glass may be due to reinforcement of the cement matrix with the apatite particles and the chemical reactivity of the apatite crystals with the acid molecules of the liquid of the luting agent [35].

V. Conclusions:

Within limitation of this study the following conclusions could be drown;

1. Formulation of GIC with sol-gel method increases its bioactivity, water sorption and solubility compared to the commercial GIC.
2. Incorporation of Nano-FAp improves the bioactivity of the sol-gel formulated GIC and decreases its water sorption.
3. Incorporation of Nano-FAp decreases solubility of sol gel formulated GIC along with increase the Nano-FAp concentration.

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