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Glass ionomer cement (GIC) is a common restorative material in dentistry, but it exhibits relatively weak mechanical properties. The present study focuses on incorporating nano-hydroxyapatite (nHAP) with different ratios (1, 3, 5, and 7wt%) in GIC to improve its properties. Mechanical properties, sorption, solubility, and diffusion coefficients after storage in distilled water for 60 days were studied. The highest sorption was measured at 7%wt (46.66 µg/mm3), and the lowest solubility was in the case of the sample containing 5% (29.166 µg/mm3). Moreover, the highest value of diffu-sion coefficient was 8.5 mm/s in the case of the sample with 7%wt nHAP. All in all, an ideal nHAP/GIC composition was prepared, and it can be applied as the basis of underneath dental filling.

Keywords

nHAP; Flexural strength; Elasticity; Diametric tensile strength; GIC

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Abstract

Glass ionomer cement (GIC) is a common restorative material in dentistry, but it exhibits relatively weak mechanical properties. The present study focuses on incorporating nano-hydroxyapatite (nHAP) with different ratios (1, 3, 5, and 7wt %) in GIC to improve its properties. Mechanical properties, sorption, solubility, and diffusion coefficients after storage in distilled water for 60 days were studied. The highest sorption was measured at 7%wt ($46.66\ \mu g/mm^3$), and the lowest solubility was in the case of the sample containing 5% ($29.166\ \mu g/mm^3$). Moreover, the highest value of diffusion coefficient was 8.5 mm/s in the case of the sample with 7%wt nHAP. All in all, an ideal nHAP/GIC composition was prepared, and it can be applied as the basis of underneath dental filling.

Keywords: nHAP, Flexural strength, Elasticity, Diametric tensile strength, GIC

1. Introduction

The concept of restoring damaged tissues and replacing lost ones using synthetic bio-materials is common, especially in dentistry. Glass ionomer cement (GIC) is a good example for reconstructive dental applications [1]. Cement is used as crowns and to fill cracks caused by tooth decay in modern dentistry instead of outdated fillings such as amalgam. Cement-based dental composites have demonstrated superior biocompatibility, natural appearance, and low plaque accumulation [2]. GIC is formed by the reaction between polyacrylic acid and calcium fluoroaluminosilicate glass powder [3—5]. Because of its effectiveness and simplicity, it is employed in medical applications, such as treating dental disorders including dental decay in the oral cavity [6,7]. Nowadays, many people suffer from tooth decay, root canal infections, and cavities which are caused by bacterial infection of the teeth and lead to the damage of the tooth structure [8]. Dental caries has become the most serious concern in oral health worldwide, owing to the potential of a carcinogenic environment forming inside the mouth cavity, which is dependent on several agents, such as diet and oral health [9]. Recently, nanometallic materials, such as Ag, TiO_2, ZrO_2, ZnO and Cu, were adopted to optimize the mechanical properties of GIC. However, these caused problems including cytotoxicity, discoloration, adhesion, and low bonding [10—14]. At the same time, several researchers began incorporating nanoparticles, nanofillers such as niobium pentoxide, bioactive glass, and forsterite, but it was found that these nanofillers have poor mechanical properties when combined with GIC, and negatively affect the release of fluoride [15—17]. Dental

materials are typically defined by evaluating mechanical parameters such as flexural strength, compressive strength, modulus of bending and others [18–21].

GIC has various advantages, such as fluoride release, transparency, and good adhesion to the tooth structure [22], as well as biocompatibility with bones, gums, and pulp. However, despite these excellent properties, its application has been restricted [23] because of its brittleness and relatively weak mechanical properties. As a result, it is used as a filling for milk teeth, to fill cracks caused by tooth decay, and as an adhesive for crowns and fractures. But it is not used for high pressure areas, such as the back teeth. To overcome these obstacles, GIC was modified by adding different materials like antibacterial compounds, silica particles, such as aluminum, hydroxyapatite (HAP), corals, eggshells, and other natural materials [6]. One of the most promising materials is HAP which has apatite-like crystal structure in the skeletal system and dental structure, as well as excellent biocompatibility. It is a promising bio-ceramic and biologically active calcium phosphate because it is similar to the bone component of humans [24]. Therefore, the effect of incorporating HAP powder into dental materials to achieve the required form and mechanical integration was studied. HAP is used as a reinforcing material and has a significant impact on the mechanical properties of polymers [24–26]. It has also been widely used as an alternative in bone and tooth repair due to its experimentally proven compatibility, as well as its ability to increase the hardness of compounds and improve surface hardness. Hydroxyapatite treated with silane added to zinc oxide was tested satisfactorily in a variety of applications [27,28]. In addition, HAP can be naturally prepared from low-cost biological sources like eggshells, seashells, and fish bones [29,30] which is highly advantageous from economic and environmental points of view. In this research, nano-HAP (nHAP) powder was prepared by calcination and added to glass cement in different proportions to study the changes in the physical and mechanical characteristics of the final material [31]. The incorporation of nHAP increases the possibility of potential applications of the developed material in dentistry [32,33].

2. Materials and methods

The powder/liquid-type GIC, Cavex Glass Ionomer from Cavex (Germany, Lot 2033134) was used. The ratio of powder to liquid was 2:1. The powder (aluminium silicate glass) consists of oxides SiO_2, Al_2O_3, B_2O_4, P_2O_5, and CaF_2, while the liquid is polyacrylic acid. nHAP (particle size <100 nm) was synthesised and characterised at the University of Basrah (Iraq) from oyster shells which were collected with dimensions in a range between 3 and 5 cm. These were purified by washing it with water and alcohol several times. Subsequently, the oyster shells were dried using an oven at 80 °C for 24 h. Then, the dried oyster shells were crushed with a pestle, and sieved with a sieve of less than 36 μm to obtain calcium carbonate. $CaCO_3$ converted to calcium oxide in an electric furnace (MV MIHM-VOGT P6/B) and then, heated up to 1200 °C for 2 h at an increment rate of 10 °C/min. After that, the sample was cooled, ground again and sieved using a 500-mesh sieve to obtain calcium oxide particles with size less than 25 μm. This was used in co-precipitation process to synthesize nHAP nanocrystals at room temperature. 2 mol of CaO was dissolved in 100 ml distilled water and 0.6 mol H_2PO_4 was added to the flask with vigorous stirring using a magnetic stirrer. The mixture was centrifuged for 10 min at 5000 rpm and washed for three times with distilled water to remove residual Ca and phosphate ions. Finally, the product was kept in an oven at 120 °C for 48 h and added in different ratios, 1%wt, 3%wt, 5% wt, and 7%wt to GIC to improve the physical and mechanical properties.

2.1. Flexural strength and modulus

For all nHAP concentrations, 24 samples ($2 \times 2 \times 25$ mm^3, Fig. 1) were prepared in a special mold according to the ISO 997-1 2007 standard. After that, the samples were stored in distilled water for 24 h at 37 °C. Flexural strengths were measured using the universal testing machine (Zwick/roll BT1-FR2.5 TN, Germany). The flexural strength (FS) in MPa was calculated using the following equation [27]:

$$FS = 3PL/(2bd^2) \tag{1}$$

where P is the maximum load exerted on the sample at the point of fracture (N), L is the distance between two supports (20 mm), b is the width (mm), while d is the thickness (mm).

Flexural modulus, E (MPa) was determined as follows [34,35]:

$$E = (3PL^3)/(4bd^3D) \tag{2}$$
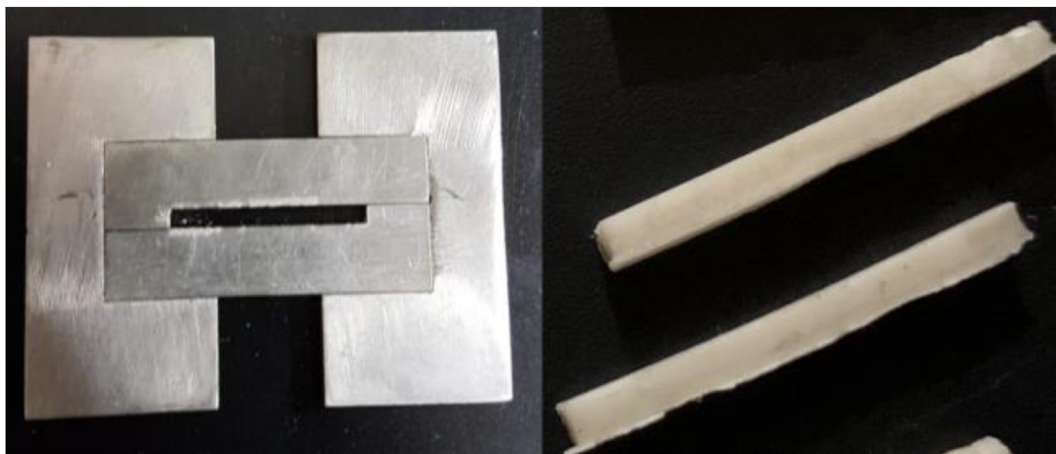
where D is the deformation of the specimen at P.

Fig. 1. Template (left) and prepared samples (right) to measure flexural strength (FS).

2.2. Diametric tensile strength (DTS)

All in all, 20 disc models (diameter 16 mm x height 10 mm, Fig. 2) were prepared to carry out diagonal tensile strength (DTS) tests. DTS was calculated by using the following equation [31]:

$$DTS\ (MPa) = 2F'/\pi RN \qquad (3)$$

where F' is the load at fracture (N), R is the sample's diameter (mm), and N is the sample's height (mm).

2.3. Water sorption and solubility

The prepared samples were stored in desiccators containing silica gel at 37 °C for one week. The samples were extracted and weighed; subsequently, the process was repeated until a constant mass was obtained which was measured (with the accuracy of ±0.1 mg (KERN ACS 220−4, Germany).

Thereafter, all samples prepared with the previously mentioned nHAP concentrations were immersed in distilled water at 37 °C. Their weights were measured at regular intervals (2 h). The weight of the samples increased on the first day in comparison to the previous day. Then, a slowdown in absorption was recorded over time until the stability of the samples was reached (mass change of less than ± 0.1 mg) (M_1). The weighing process continued for 60 days, after which the samples were extracted from distilled water and placed into the silica gel containing desiccator and weighed a week later (M_2). Subsequently, the solubility (WS) and absorbance (WA) were calculated according to the glass cement (Cave, Germany, Lot 2033134 ISO 9917-1:2007) standard using the following equations [27,35]:

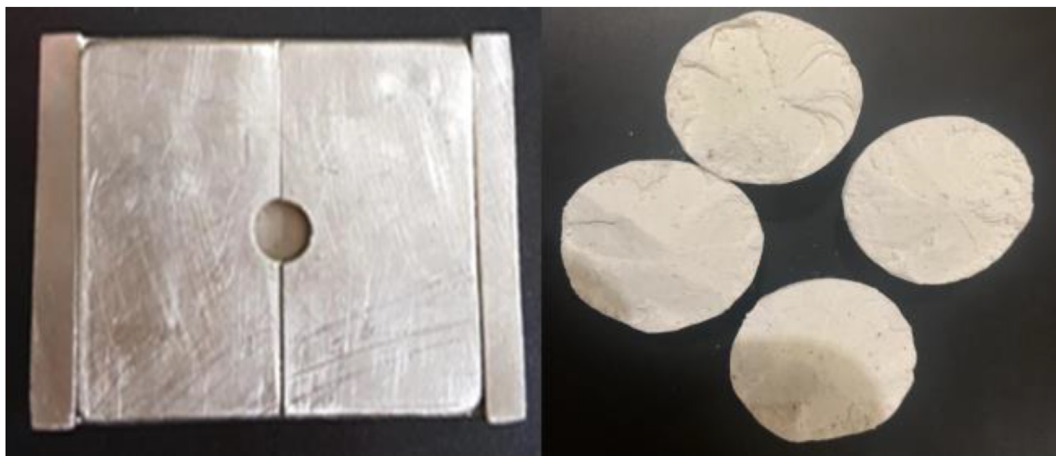$$WA = (M_1 - M_2)/V \qquad (4)$$

$$WS = (M_0 - M_2)/V \qquad (5)$$

Fig. 2. Template (left) and prepared samples to measure diametric tensile strength (DTS).

where V is the volume.

2.4. Diffusion coefficients

To obtain the diffusion coefficient by using Fick's second law the Stefan approximation was applied as follows:

$$\frac{\partial c}{\partial t} = D\left(\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2} + \frac{\partial^2 c}{\partial z^2}\right) \tag{6}$$

where t (s) is time, and c (%) is the concentration. The diffusion coefficient of the 1D model for the mass flow in the solids is D (ms^{-1}), and the differential equation is expressed in the following form:

$$\frac{\partial c}{\partial t} = D\left(\frac{\partial^2 c}{\partial x^2}\right) \tag{7}$$

The solution of Fick's second law for longer periods of diffusion is given [28] as follows:

$$\frac{M_t}{M_\infty} = -\frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp\left[\frac{(2n+1)^2 \pi^2 D t}{L^2}\right] \tag{8}$$

and it is reduced by the initial stage of absorption when the value of $M_t/M_\infty \leq 0.6$.

$$\frac{M_t}{M_\infty} = \frac{4}{L}\left(\frac{Dt}{\pi}\right)^{\frac{1}{2}} \tag{9}$$

In eq. (9), the diffusion coefficient of the liquid during the adsorption is D (mm/s), and M_t expresses the absorption at time t (s), while M_∞ is the mass (g) of absorption when the weight is constant, and L is the thickness of the sample.

If the uptake M_t is determined at reasonable time intervals till the equilibrium state is attained, then subsequently, the graph of M_t/M vs $t^{1/2}$ should show a straight line, and the slope of the line is S which can also be calculated by the following equation [36]:

$$S = 4\left(\frac{D}{\pi L^2}\right)^{\frac{1}{2}} \tag{10}$$

3. Results and discussion

Samples consisting of different ratios of nHAP mixed with GIC were prepared to enhance mechanical and physical properties of the latter. After 60 days of storage in distilled water, the characteristics of the prepared nHAP/GIC materials were investigated, and flexural strength (FS), modulus diametric tensile strength (DTS), diffusion,

Table 1. Flexural strength (FS) values (in MPa) of the pure glass ionomer cement (GIC) without nano-hydroxyapatite (nHAP) and the prepared nHAP/GIC samples with different weight ratios of nHAP.

Weight ratios of nHAP (%)	Flexural strength (Mpa)±SD
0	24.794 ± 3.71
1	24.898 ± 3.38
3	26.796 ± 0.82
5	29.951 ± 2.72
7	13.446 ± 0.69

solubility and absorption coefficient measurements were performed. The flexural strength of the samples ranged from 13.446 MPa to 29.951 MPa, where the lowest value belongs to the one with 7 wt% nHAP (Table 1). This decrease in FS is attributed to the increase in nHAP content, which leads to a more brittle structure due to the lack of complete powder-liquid bonding. The highest FS value (29.951 MPa) corresponds to the sample which contain 5wt% nHAP, and it is better than the GIC (Cavex, Germany, Lot 2033134 ISO 9917-1:2007) standard which is used as reference to compare the results. The incorporation of nHAP enhanced the external strength compared to pure GIC [30]. The increase in the surface area caused by nHAP and its good diffusion in the powder increased the bonding, which provided more flexibility. HAP porosity enhances the amount of external strength of GIC [37].

The flexural modulus was also measured (Table 2). The flexural modulus increased by the increasing amount of nHAP up to 5wt%, but it dropped to even below the pure GIC at 7 wt%. The flexural modulus values ranged between 175.94 MPa and 742.96 MPa, and the largest value belonged to 5wt% (742.96 MPa), whereas the lowest one corresponded to the sample with 7wt% at 175.94 MPa.

The diametric tensile strength (DTS) of the ranged from 7.52 MPa to 14.01 MPa (Table 3). The highest and lowest values were 5 wt% and 7 wt% with 7.52 and 14.01 MPa, respectively. Because the nHAP filled the empty spaces, the integration of nHAP into GIC boosted the DTS by up to 5% wt compared to pure GIC. Thus, the formation of cracks was

Table 2. Flexural modulus (MPa) values of the pure glass ionomer cement (GIC) without nano-hydroxyapatite (nHAP) and the prepared nHAP/GIC samples with different weight ratios of nHAP.

Weight ratios of nHAP (%)	Flexural Modulus (MPa) ± SD
0	176.84 ± 0.95
1	317.46 ± 1.25
3	428.94 ± 1.31
5	742.96 ± 0.85
7	175.94 ± 1.35

Table 3. Diametric tensile strength (DTS) values (MPa) of the pure glass ionomer cement (GIC) without nano-hydroxyapatite (nHAP) and the prepared nHAP/GIC samples with different weight ratios of nHAP.

Weight ratios of nHAP (%)	Diametric tensile strength (MPa) ±SD
0	13.05 ± 0.64
1	12.23 ± 0.77
3	12.96 ± 0.35
5	14.01 ± 0.39
7	7.52 ± 0.46

prevented in case of pores and blemishes. However, further increase of nHAP content (7%) led to brittle nHAP/GIC material.

The sample with 7wt% nHAP showed a significant change in DTS as well compared to the others and having even less suitable properties than pure glass cement. This decrease is attributed to the preponderance of nHAPs overwhelmed by the reaction with polyacrylic acid. In addition, agglomeration of nHAP may result in heterogeneous dispersion in the matrix. Generally, GIC is water sensitive, and it has a high solubility in the sitting process (processing process), probably decreasing the mechanical properties [38].

The water sorption of the nHAP/GIC samples was also determined (Fig. 3). The values were computed for all ratios after being immersed in water for different periods of time (1, 7, and 60 days). The results indicated that the values of the water sorption increased when the immersion period in water increased. The sorption values ranged from 0 to 50 $\mu g/mm^3$. The highest value of sorption was 46.66 ± 2.08 $\mu g/mm^3$ at 60 days experienced in case of the sample with 7wt% nHAP, whereas the lowest value was 34.66 ± 2.08 $\mu g/mm^3$ which is corresponding to the sample with 5wt% nHAP. One of the weaknesses of GIC is that it is hydrolytically
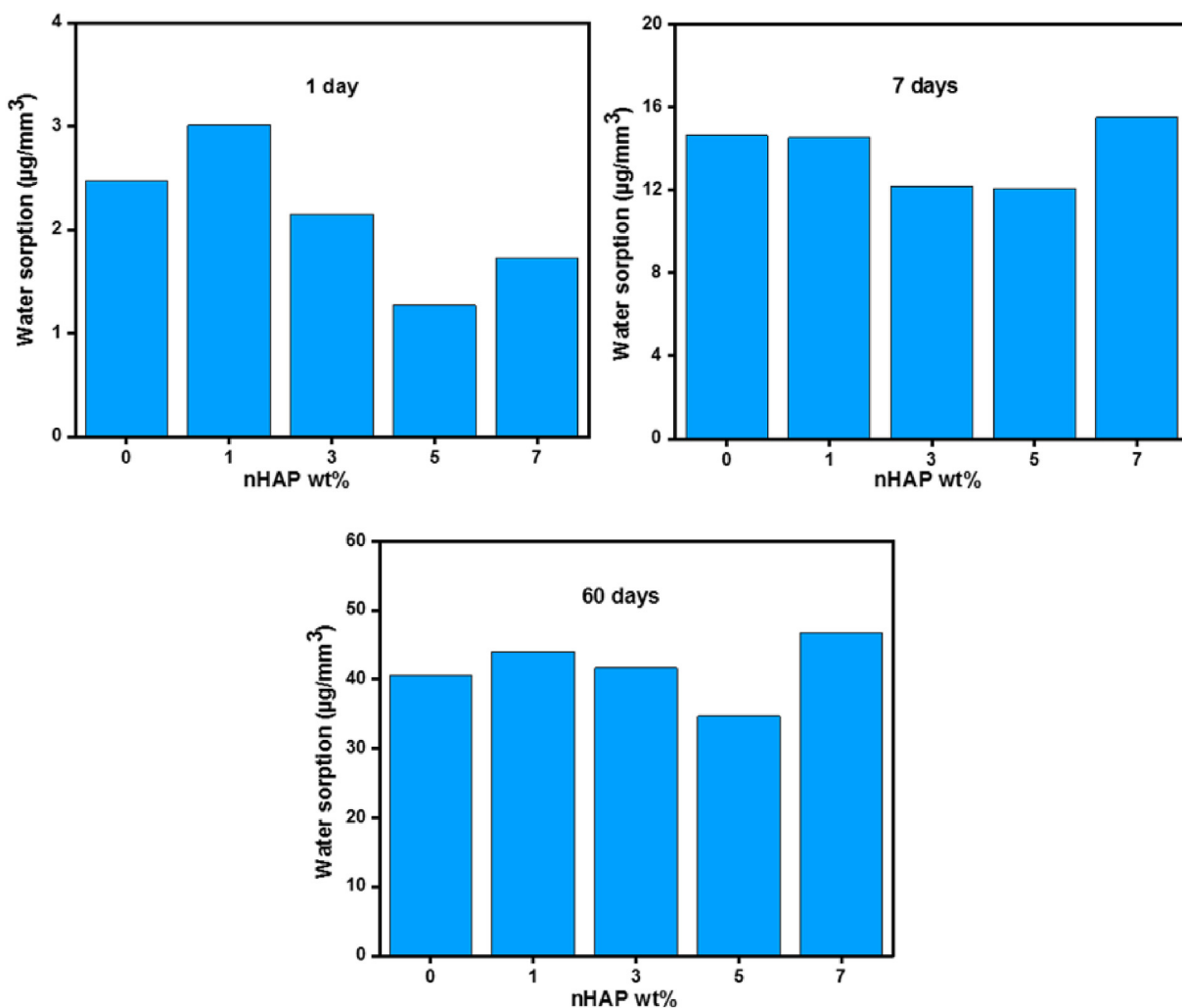

Fig. 3. Water sorption ($\mu g/mm^3$) of the pure GIC and the prepared nHAP/GIC samples (0, 1, 3, 5, and 7 wt% of nHAP) during different periods of times (1, 7, and 60 days).
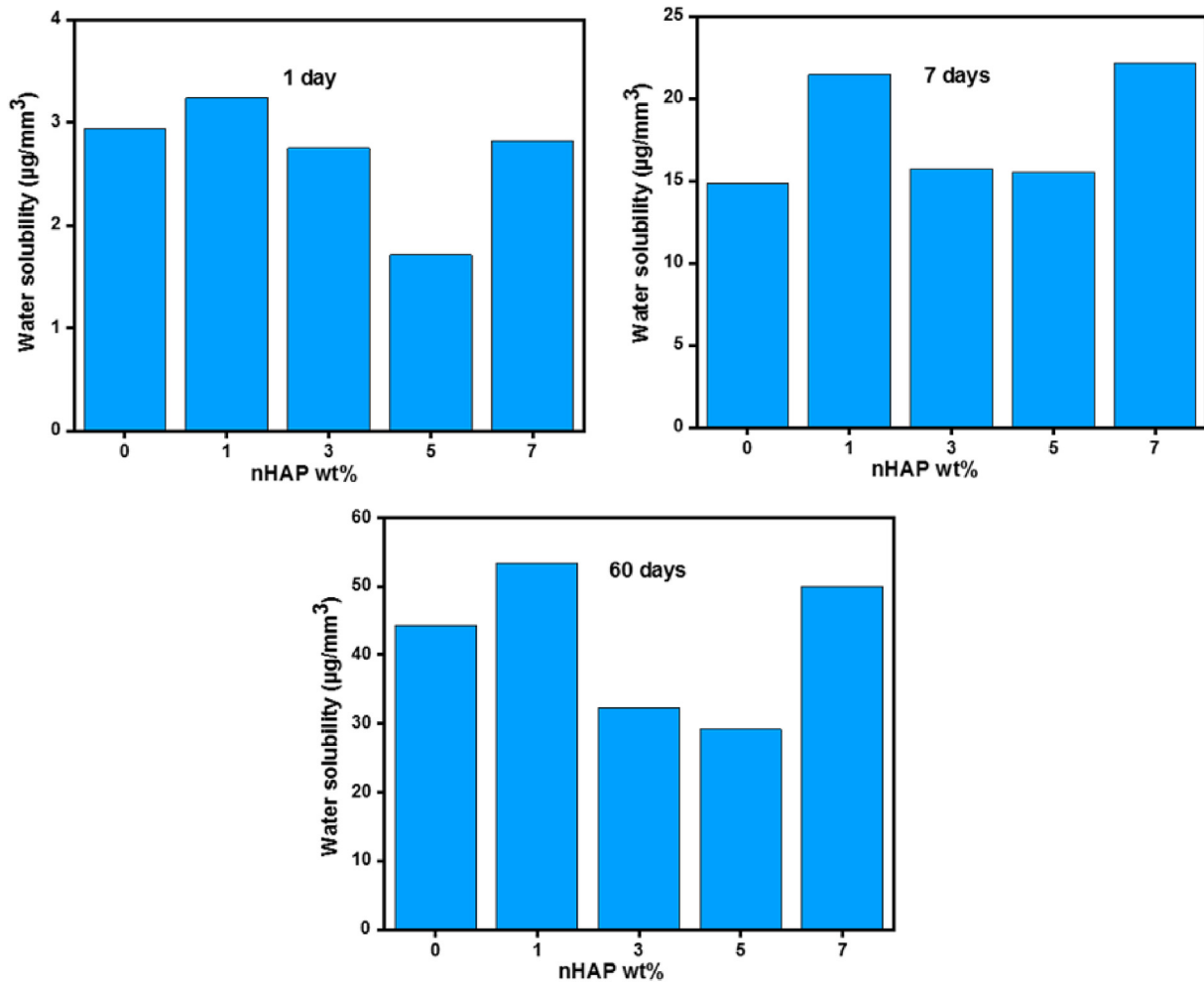
Fig. 4. Water solubility ($\mu g/mm^3$) of the modified GIC mixing with various ratios (0, 1, 3, 5 and 7) wt% from nHAP during different periods (1, 7 and 60 days).

susceptible at initial stages of setting when exposed to air and moisture. It was previously reported that water sorption and solubility is one of the most critical attributes for a restorative material [39]. In addition, the increase in sorption and solubility can be linked with surface roughness.

The water solubility of the samples was determined (Fig. 4). It was found that the solubility ranges from 0 to 60 $\mu g/mm^3$. The highest solubility was at 60 days (1%) and was equal to 53.66 ± 5.06 $\mu g/mm^3$, whereas the lowest value was at 5wt% and equal to 29.17 ± 2.02 $\mu g/mm^3$.

The solubility of the modified GIC samples, without statistical differences at first day is due to the incomplete preparation and unproduced ammonium polyacrylate. The difference appeared in the measurements of 1 and 60 days, and the samples with 1 wt% and 7 wt% nHAP content had higher solubility while those with 3wt% and 5wt% nano-hydroxyapatite showed a statistically

Table 4. Diffusion coefficient (mm/s) of the pure GIC and the prepared nHAP/GIC samples (0, 1, 3, 5, and 7 wt% of nHAP).

Weight ratios of nHAP %	Diffusion coefficient (mm/s) ± SD
0	4.6 ± 1.52
1	3.4 ± 1.15
3	3.6 ± 1.21
5	4.2 ± 1.41
7	8.5 ± 2.84

significant decrease in solubility. This finding could be associated with the improved microstructure of the modified GIC by addition of a certain amount of nHAP.

The diffusion coefficients of the modified GIC with different ratios (0, 1, 3, 5, and 7wt%) of nHAP were determined (Table 4). The results shows that the diffusion coefficient ranges from 3.4 mm/s to 8.5 mm/s, and the highest and lowest values belong

to the sample with the highest nHAP content and lowest nHAP content, respectively.

Thus, with increasing nHAP added to GIC, its sitting time increases. At all concentrations, the siting time remained in the rational range of 90−360 s which is regulated in ISO:9917-1, except in case of the sample with 7wt% nHAP content, which surpasses the uppermost limit. The best concentricity of nHAP was indicated at 5wt% by weight because it could provide dentists with a more convenient time to mix powder and liquid. It also has better physical and clinical properties for absorbance and solubility than other concentrations.

4. Conclusion

Nano-hydroxyapatite prepared in our laboratories from oyster shells was successfully added to glass ionomer cement in four different formulations (1, 3, 5, and 7 wt.%). nHAP/GIC samples were tested and compared to pure glass. Incorporation of a properly selected amount of nHAP makes GIC useful for significant restorative application in areas subject to high compressive forces. For all concentrations, the positioning time remained in the logical range between 90 and 360 s regulated in ISO:9917-1, except in the case of the 7 wt% sample, which is above the upper limit. An improvement was also observed in the physical properties of a 5% wt sample in terms of absorption and solubility. With these results, we observed a greater improvement in mechanical and physical properties compared to pure GIC in the case of the sample with 5 wt% nHAP content according to ISO: 9917-1. All in all, an ideal cement formulation has been successfully developed. It has been found that this nHAP/GIC can be applied as a basis under dental fillings, because its properties fulfil the recommendations of the American Dental Association (ADA).

Acknowledgement

References

[1] N. Shariq, Z. Khurshid, M.S. Zafar, A.S. Khan, S. Zohaib, J.M.N. Martí, S. Sauro, J.P. Matinlinna, I. Rehman, Modifications in glass ionomer cements: nano-sized fillers and bioactive nanoceramics, Int. J. Mol. Sci. 17 (2016) 1134, https://doi.org/10.3390/ijms17071134.

[2] M. Anoj, D. Bisht, R. Yadav, S. Saini, G.S. Dangayach, A. Patnaik, M.L. Meena, Fabrication and characterization of micro alumina zirconia particulate filled dental restorative composite materials, Polym. Compos. 43 (2022) 1526−1535, https://doi.org/10.1002/pc.26473.

[3] M. Alireza, S. Ansari, M. Moshaverinia, N. Roohpour, J.A. Darr, I. Rehman, Effects of incorporation of hydroxyapatite and fluoroapatite nanobioceramics into conventional glass ionomer cements (GIC), A Biomater. 4 (2008) 432−440, https://doi.org/10.1016/j.actbio.2007.07.011.

[4] M.Z.A. Khiri, K.A. Matori, M.H.M. Zaid, A.C. Abdullah, N. Zainuddin, W.N.W. Jusoh, R.A. Jalil, N.A.A. Rahman, E. Kul, S.A.A. Wahab, N. Effendy, Soda lime silicate glass and clam shell act as precursor in synthesize calcium fluoroaluminosilicate glass to fabricate glass ionomer cement with different ageing time, J. Mater. Res. Technol. 9 (2020) 6125−6134, https://doi.org/10.1016/j.jmrt.2020.04.015.

[5] I.A. Rahman, S.M. Masudi, N. Luddin, R.A. Shiekh, One-pot synthesis of hydroxyapatite−silica nanopowder composite for hardness enhancement of glass ionomer cement (GIC), B Mater. Sci. 37 (2014) 213−219, https://doi.org/10.1007/s12034-014-0648-3.

[6] R. Liu, E. Wang, Y. Guo, Q. Zhou, Y. Zheng, J. Zhai, K. Zhang, B. Zhang, Enhanced antibacterial properties and promoted cell proliferation in glass ionomer cement by modified with fluorinated graphene-doped, J. Appl. Biomer. Func. Mater. 19 (2021) 22808000211037487, https://doi.org/10.1177/22808000211037487.

[7] Z. Keshi, L. Zheng, J. Xing, S. Chen, R. Chen, L. Ren, Mechanical, antibacterial, biocompatible and microleakage evaluation of glass ionomer cement modified by nano-hydroxyapatite/polyhexamethylene biguanide, Dent. Mater. J. (2022) 2021−2096, https://doi.org/10.4012/dmj.2021-096.

[8] R. Yadav, M. Kumar, Dental restorative composite materials: a review, J. Oral Biosci. 61 (2019) 78−83, https://doi.org/10.1016/j.job.2019.04.001.

[9] Y. Liu, T. Kohno, R. Tsuboi, P. Thongthai, D. Fan, H. Sakai, H. Kitagawa, S. Imazato, Antibacterial effects and physical properties of a glass ionomer cement containing BioUnion filler with acidity-induced ability to release zinc ion, Dent. Mater. J. 40 (2021) 1418−1427, https://doi.org/10.4012/dmj.2021-052.

[10] K.R. Kantovitz, F.P. Fernandes, I.V. Fernandes, M.O. Lazzarini, G.C. Denucci, O.P. Gomes, P.A. Giovani, K.M.S. Moreira, V.G.A. Pecorari, A.F.S. Borges, F.H. Nociti, R.T. Basting, P.N. Lisboa, R.M.P. Rontani, TiO2 nanotubes improve physico-mechanical properties of glass ionomer cement, Dent Mater 36 (2020) e85−e92, https://doi.org/10.1016/j.dental.2020.01.018.

[11] V.V.A. Thampi, M. Prabhu, K. Kavitha, P. Manivasakan, P. Prabu, V. Rajendran, S. Shankar, P. Kulandaivelu, Hydroxyapatite, alumina/zirconia, and nanobioactive glass cement for tooth-restoring applications, Ceram. Int. 40 (2014) 14355−14365, https://doi.org/10.1016/j.ceramint.2014.06.027.

[12] I.A. Rahman, N.A.M. Ghazali, W.Z.W. Bakar, S.M. Masudi, Modification of glass ionomer cement by incorporating nanozirconia-hydroxyapatite-silica nano-powder composite by the one-pot technique for hardness and aesthetics improvement, Ceram. Int. 43 (2017) 13247−13253, https://doi.org/10.1016/j.ceramint.2017.07.022.

[13] D.A. Perez, R.V. Coronado, J.M.U. Cervantes, N.R. Fuentes, C. Aparicio, C. Covarrubias, M.A. Perez, V.G. Perez, M.M. Hernandez, J.V.C. Rodriguez, Antibacterial activity of a glass ionomer cement doped with copper nanoparticles, Dent. Mater. J. (2020) 2019−2046, https://doi.org/10.4012/dmj.2019-046.

[14] R.M. Al Badr, Investigating some of the mechanical properties of glass-ionomers cements modified with ZnO and TiO2 Nanoparticles, D J P Sci. 15 (2019) 85−97, https://doi.org/10.24237/djps.15.04.503A.

[15] F.S. Sayyedan, M.H. Fathi, H. Edris, A. Doostmohammadi, V. Mortazavi, A. Hanifi, Effect of forsterite nanoparticles on mechanical properties of glass ionomer cements, Ceram. Int. 40 (2014) 10743−10748, https://doi.org/10.1016/j.ceramint.2014.03.062.

[16] J. Sasaki, W. Kiba, G.L. Abe, C. Katata, M. Hashimoto, H. Kitagawa, S. Imazato, Fabrication of strontium-releasable inorganic cement by incorporation of bioactive glass, Dent. Mater. 35 (2019) 780–788, https://doi.org/10.1016/j.dental.2019.02.019.

[17] I.M. Garcia, V.C.B. Leitune, G.D.S. Balbinot, S.M.W. Samuel, F.M. Collares, Influence of niobium pentoxide addition on the properties of glass ionomer cements, A Biomer. Odon. Scand. 2 (2016) 138–143, https://doi.org/10.1080/23337931.2016.1239182.

[18] R. Murugan, F. Yazid, N.S. Nasruddin, N.N.M. Anuar, Effects of nanohydroxyapatite incorporation into glass ionomer cement (GIC), Minerals 12 (2021) 9, https://doi.org/10.3390/min12010009.

[19] R. Yadav, H.H. Lee, Ranking and selection of dental restorative composite materials using FAHP-FTOPSIS technique: an application of multi criteria decision making technique, J. Mech. Behav. Biomed. Mater. 132 (2022) 105298, https://doi.org/10.1016/j.jmbbm.2022.105298.

[20] R. Yadav, A. Meena, Mechanical and two-body wear characterization of micro-nano ceramic particulate reinforced dental restorative composite materials, Polym. Compos. 43 (2022) 467–482, https://doi.org/10.1002/pc.26391.

[21] R. Yadav, A. Meena, Effect of aluminium oxide, titanium oxide, hydroxyapatite filled dental restorative composite materials on physico-mechanical properties, Ceram. Int. 48 (2022) 20306–20314, https://doi.org/10.1016/j.ceramint.2022.03.311.

[22] A.A. Ashour, S. Basha, N.H. Felemban, E.T. Enan, A.A. Alyamani, S.M.F.G. El-Rab, Antimicrobial efficacy of glass ionomer cement in incorporation with biogenic zingiber officinale capped silver-nanobiotic, chlorhexidine diacetate and lyophilized miswak, Molecules 27 (2022) 528, https://doi.org/10.3390/molecules27020528.

[23] S.D. Aldhuwayhi, A. Sajjad, W. Zaripah W. Bakar, D. Mohamad, T.P. Kannan, I.A. Moheet, Evaluation of fracture toughness, color stability, and sorption solubility of a fabricated novel glass ionomer nano zirconia-silica-hydroxyapatite hybrid composite material, Int. J. Poly. Sci. 2021 (2021) 8, https://doi.org/10.1155/2021/6626712.

[24] S.C. Wu, Y.L. Kao, Y.C. Lu, H.C. Hsu, W.F. Ho, Preparation and characterization of microrod hydroxyapatite bundles obtained from oyster shells through microwave irradiation, J. Aust. Ceram. Soc. 57 (2021) 1541–1551, https://doi.org/10.1007/s41779-021-00657-3.

[25] S. Rujitanapanich, P. Kumpapan, P. Wanjanoi, Synthesis of hydroxyapatite from oyster shell via precipitation, Energy Proc. 56 (2014) 112–117, https://doi.org/10.1016/j.egypro.2014.07.138.

[26] R. Yadav, A. Meena, Comparative investigation of tribological behavior of hybrid dental restorative composite materials, Ceram. Int. 48 (2022) 6698–6706, https://doi.org/10.1016/j.ceramint.2021.11.220.

[27] R.M. AlBadr, S.A. Halfi, K.M. Ziadan, The effectiveness of oyster filler on the physical and mechanical properties of novel dental restorative composite, in: AIP Con. Proc, AIP Publishing LLC. 2020 050001, https://doi.org/10.1063/5.0031467 vol. 2290.

[28] R. Yadav, M. Kumar, Investigation of the physical, mechanical and thermal properties of nano and microsized particulate-filled dental composite material, J. Compos. Mater. 54 (2020) 2623–2633, https://doi.org/10.1177/0021998320902212.

[29] X.Y. Lü, Y.B. Fan, D. Gu, W. Cui, Preparation and characterization of natural hydroxyapatite from animal hard tissues, in: Key Eng. Mater., Trans Tech Publications Ltd.. 2007, pp. 213–216, https://doi.org/10.4028/www.scientific.net/KEM.342-343.213, vol. 342.

[30] N.A.S.M. Pu'ad, P. Koshy, H.Z. Abdullah, M.I. Idris, T.C. Lee, Syntheses of hydroxyapatite from natural sources, Heliyon 5 (2019) e01588, https://doi.org/10.1016/j.heliyon.2019.e01588.

[31] A.D. Wilson, Glass-ionomer Cement, the Setting Reaction and its Clinical Consequences, 1988.

[32] R. Yadav, Fabrication, characterization, and optimization selection of ceramic particulate reinforced dental restorative composite materials, Polym. Compos. 30 (2022) 09673911211062755, https://doi.org/10.1177/09673911211062755.

[33] R. Yadav, A. Meena, A. Patnaik, Tribological behavior of zinc oxide-hydroxyapatite particulates filled dental restorative composite materials, Polym. Compos. 43 (2022) 3029–3040, https://doi.org/10.1002/pc.26597.

[34] M. Atai, A. Pahlavan, N. Moin, Nano-porous thermally sintered nano silica as novel fillers for dental composites, Dent. Mater. 28 (2012) 133–145, https://doi.org/10.1016/j.dental.2011.10.015.

[35] R.M. Al-Bader, K.M. Ziadan, M.S. Al-Ajely, Water adsorption characteristics of new dental composites, Int. J. Med. Res. Health Sci. 4 (2015) 281–286, https://doi.org/10.5958/2319-5886.2015.00052.1.

[36] M.M. Basir, M. Ataei, M. Rezvani, Effect of incorporation of various amounts of nano-sized hydroxyapatite on the mechanical properties of resin modified glass ionomer, J. Dent. Sch. Shahid Beheshti Univ. Dental J. 30 (2011) 216–223. http://research.shahed.ac.ir/WSR/SiteData/PaperFiles/8106_900625236.pdf.

[37] K. Lin, J. Chang, R. Cheng, M. Ruan, Hydrothermal microemulsion synthesis of stoichiometric single crystal hydroxyapatite nanorods with mono-dispersion and narrow-size distribution, Mater. Lett. 61 (2007) 1683–1687, https://doi.org/10.1016/j.matlet.2006.07.099.

[38] C.R. Brito, L.G. Velasco, G.A.V.C. Bonini, J.C.P. Imparato, D.P. Raggio, Glass ionomer cement hardness after different materials for surface protection, J. Biomed. Mater. Res. Part A 93 (2010) 243–246, https://doi.org/10.1002/jbm.a.32524.

[39] G. Wang, B.M. Culbertson, D. Xie, R.R. Seghi, Physical property evaluations of perfluorotriethylene glycol dimethacrylate as a potential reactive diluent in dental composite resins, J. Macr. Sci. Part A 36 (1999) 225–236, https://doi.org/10.1081/MA-100101527.