Sol-Gel Synthesis of Soda Lime Silica-Based Bioceramics Using Waste as Renewable Sources

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Research Article

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DOI: https://doi.org/10.21203/rs.3.rs-703473/v1

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Sol-gel synthesis of soda lime silica-based bioceramics using waste as renewable sources

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Abstract

The purpose of the work is to prepare and assess soda lime silica-based (SiO$_2$-CaO-Na$_2$O) bioactive ceramics using waste as renewable sources. Thus we produced a SiO$_2$-CaO-Na$_2$O-based bioactive ceramic by sol-gel process using rice husk and eggshells as sources of silica and calcium oxide, respectively. The precursors such as calcinated eggshell powder, rice husk ash (RHA) and sodium hydroxide (NaOH) were processed by the sol-gel method, resultant in SiO$_2$-CaO-Na$_2$O-based bioactive ceramics. The gel derived sintered sample showed combeite high (Na$_6$Ca$_3$Si$_6$O$_{18}$) as a major crystalline phase. Subsequently, the sintered specimens were analyzed from the physical and structural point of view, and in terms of apatite mineralization rate in simulated environments and cytocompatibility in relative to human osteoblast-like cells. The studies showed that the produced crystalline SiO$_2$-CaO-Na$_2$O-based ceramics showed an average porosity of 45%. In vitro evaluation of the biological properties revealed that the prepared ceramics possesses the mineralization of carbonated hydroxyapatite (CHA) in a simulated environment with good cytocompatibility and controlled degradation rate. Therefore, the results obtained suggest that the prepared SiO$_2$-CaO-Na$_2$O-based bioactive ceramics using waste as renewable sources might be a low cost ceramics for applications in biomedical field.
Keywords: Bioceramics, renewable sources, bioactivity, cytocompatibility, biomedical application

Introduction

Bioceramics are defined as the synthetic crystalline inorganic material used for the healing and replacement of injured or diseased parts of the human tissue. When bioceramics are implanted in body tissue defects, the complex chemical and biological process occurs immediately due to the interaction between bioceramics and living cells that determines the progression of the tissue regeneration process [1, 2]. Several bioceramics have been explored to date as potential candidates for tissue engineering applications starting from alumina (Al₂O₃) and zirconia (ZrO₂) [3, 4] to calcium phosphates and calcium silicates owing to their exceptional osteoconductivity and biocompatibility [5, 6]. Wide variety of bioceramics have been designed based on the need and applications, comprised of thin layer coating on metallic implants, microspheres, composites by combined with biopolymer materials, large well-polished surfaces and porous networks [7]. Furthermore, loading of bioceramic composite scaffolds with drugs for therapeutic purposes such as antibacterial and anti-tumor effects has also been explored [8]. However, due to the lack of quality of food structure with increasing population, bone defects have become common diseases, resulting in increasing clinical demands on tissue regeneration and wound healing. Therefore, improvement of new fabrication procedures of bioceramics is a significant aspect that researchers are exploring and trying to point out.

In general, conventional solid state sintering, melt quenching subsequent crystallization and sol-gel technique have proven to be most commonly used preparation techniques for bioceramics [9, 10]. Solid state sintering and melting methods require high temperature processing; during high temperature, evaporation of the volatile component is a
drawback of these methods. Whereas, sol-gel technique requires low processing temperatures and it provides high chemical homogeneity [11, 12]. This method allows a wide variety of compositions with the production of different shapes like powders, monoliths, coatings or fibers [13]. Moreover, bioceramics obtained from sol-gel technique shows porosity and higher surface areas which are favourable factors for the bioactivity [14, 15].

The conventional sol-gel process requires high purity alkoxy silane precursors such as tetramethylorthosilicate (TMOS) and tetraethylorthosilicate (TEOS) as silica precursors and calcium nitrate tetra hydrate (Ca(NO$_3$)$_2$.4H$_2$O) as calcium oxide precursor. Nevertheless, these materials are costly. Rice husk as a silica source and eggshells as a calcium oxide source could be low-cost raw materials for sol–gel processing of bioceramics. Therefore, in the present study, a sol–gel technique was employed to produce soda lime silica-based (SiO$_2$:CaO:Na$_2$O) bioceramics using silica source of rice husk and calcium oxide source of eggshells with the objective of providing a method for producing cost effective soda lime silica-based bioceramics for applications in biomedical field.

2. Materials and methods

2.1. Preparation of bioceramics

The bioceramic with stoichiometric balance of chemical composition 50% SiO$_2$-25% CaO-25% Na$_2$O (mol%) was produced by sol-gel process using calcinated eggshell powder, rice husk ash (RHA) and sodium hydroxide (NaOH; 99.9% purity, Sigma Aldrich). Eggshells and rice husk were collected from NIT canteen and local rice mill, respectively. Rice husk was cleaned using diluted hydrochloric acid as per the procedure proposed by Abu Bakar et al [16]. The washed rice husk was then calcination at 600 °C for 4 h to achieve high purity amorphous silica (~ 99%) as RHA. Boiled chicken eggshells were washed using distilled water and then dried. Afterwards, the eggshells were made into fine powder. High purity
calcium oxide (~ 99%) was obtained by the calcination of fine powder of eggshells at 900 °C for 2 hours. The sol preparation was comprised of mixing of stoichiometric amount of RHA in 1M NaOH solution. Thus sodium silicate solution was accomplished by simultaneous boiling and stirring of RHA in NaOH solution for 30 min at 60 °C. 2M nitric acid was mixed with calcinated eggshell powder, resulting in a calcium nitrate solution. Sodium silicate solution was added drop wise into the solution containing calcinated eggshell powder and nitric acid under continuous stirring to produce white gel. The obtained white gel was kept for 3 days for aging at 70 °C and then dried for 6 h at 150 °C. The block of the solid mass of the dried gel was made into fine powder and then calcinated at 700 °C for 2 h. The calcinated powder was made into pellets under a pressure of about 80 MPa using poly vinyl alcohols (PVA) as a binder. The sintering of resultant pellets was carried out at 900 °C for 2 hours according to the TG-DTA analysis to achieve optimum crystalline ceramics.

2.2. Characterization

The typical calcination and sintering temperatures were estimated from the simultaneous thermogravimetric (TG) and differential thermal analysis (DTA) (Model: NETZSCH, Germany) of dried gel powder sample. The phase identification of the synthesized bioceramic samples was carried out by means of powder X-ray diffractometer (XRD) (Model: PANALYTICAL XPERT POWDER) using CuKα, Ni filtered radiation with 0.02 step size and time per step 50 s. Fourier transform infrared (FTIR) spectroscopy (model S 100; PerkinElmer) was carried out to study the molecular arrangement of the samples. The infrared spectrum was recorded in transmittance mode in a wavelength range of 400 – 4000 cm⁻¹ and with a resolution of 4 cm⁻¹ using KBr pelleted samples. Microstructural images along with elemental configurations were studied by scanning electron microscopy (SEM) jointly equipped with energy dispersive spectroscopy (EDS) (model 5WEGA 3 LMU; TESCAN).
2.3. Physical properties

The percentage of linear shrinkage (LS) of ceramics was determined by measuring the changes in diameter of the specimens before and after sintering using equation (1). The relative density ($\rho$) and open porosity ($P_o$) were determined as per the ASTM B962-17 by using Archimedes' principle [17] from equations (2&3).

$$LS\% = \frac{l_1-l_0}{l_0} \times 100$$

(1)

$$\rho = \frac{\rho_a \times 100}{\phi}$$

(2)

$$P_o = \frac{W_3-W_1}{W_3-W_2} \times 100$$

(3)

Where $l_0$ is the diameter of the specimen before sintering, $l_1$ is diameter of the specimen after sintering, $\rho_a$ is apparent density, $\phi$ is theoretical density of the material, $W_1$ is weight of sintered specimen in air, $W_2$ is weight of specimen suspended in deionized water and $W_3$ is the weight of specimen after being saturated in deionized water.

2.4. In vitro bioactivity

Simulated body fluid (SBF) test was developed as a primary test to evaluate the in vitro bioactivity of the biomaterials. The test was conducted according to the ISO 10993-14. SBF solution was made in consistent with the process described by Kokubo et al. [18, 19]. In order to examine the apatite layer mineralization rate, sintered pellets (3 mm thickness and 10 mm diameter) at 900 °C were ultrasonically rinsed, left to air dry and then placed in torsion beakers containing SBF solution for various time periods at 37 °C. The assessment was carried out under static condition to observe the change in pH level of SBF solution over immersion period. After the prearranged soaking time, the pellets were taken out from the
beakers, rinsed with deionised water and then dried. The change in surface microstructure, chemical composition, molecular arrangement and crystalline phase were studied using SEM, EDS, FTIR and XRD, respectively.

2.5. In vitro degradation

Degradation behavior of synthesized SiO$_2$-CaO-Na$_2$O-based ceramics was studied under two different physical conditions, such as SBF and Tris-HCl buffer solutions. The test was performed on $\varnothing$ 10 mm and 3 mm thickness ceramic discs. The initial weights of the samples were noted and were then transferred into 50 ml torsion centrifuge tubes containing SBF and Tris-HCl buffer solutions. The volume of the solution was calculated using the principle $V_s = S_a/10$, where $S_a$ is the surface area of the pellets and $V_s$ is the volume of the solution. The immersed specimens were maintained at 37 °C for various time durations (1, 3, 7, 14 and 21 days) and the solution was exchanged every 3 days. At the end of each soaking point, the discs were removed from the medium, rinsed with distilled water and then dried at 90 °C. The change in weight loss was measured and the degradation was calculated according to the percentage of original weight. The experiment was conducted in triplicate for each soaking point and the results were expressed as mean ± SD.

2.6. Cytocompatibility test

The cellular behaviour and cytocompatibility potential of prepared bioceramics were investigated using MG-63 cells by *in vitro* biological assay. Viability of cells in the presence of bioceramics was measured using colorimetric MTT [3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyltetrazoliumbromide] assay. Dulbecco Modified Eagle’s Medium (DMEM) accompanied with 10% fetal bovine serum (FBS) was used as culture medium. After 24 h of seeding the MG-63 cell in a 96 wells plate ($5 \times 10^3$ cells per well), specific concentrations of bioceramic particle were added into each well and incubated at 37 °C humidified air with 5%...
CO$_2$ for 48 h. 20µL (5 mg/mL) MTT reagent was added into each well and incubated at 37 °C for 4 h. The culture medium with MTT of all wells was removed by pipetting it out and then 100 µL of acidified isopropanol was added to each well followed by incubation for half an hour to solubilize the formazan crystals. In order to measure the optical density microplate reader was used (wavelength 570 nm). The percentage of cell viability was calculated by the following formula:

$$\text{%Cell Viability} = \frac{[OD]_{\text{test}}}{[OD]_{\text{control}}} \times 100$$ (1)

3. Results and discussion

3.1. TG-DTA analysis

Fig. 1 demonstrates the simultaneous thermogravimetric (TG) and differential thermal analysis (DTA) of dried powder sample. Thermogravimetric measurement curve showed primarily three distinctive stages of mass reduction. The initial stage was observed in the temperature region of 30 – 150 °C with the mass reduction of ~ 5.3% and it was appeared in DTA measurement as a small endothermic shoulder at ~ 88 °C, corresponded to the elimination of adsorbed water [20]. The next stage of mass reduction which accomplished in the temperature region of 150 – 480 °C, exhibited an endothermic peak at ~ 270 °C and a small mass reduction of ~ 5.7%, which was related to chemically adsorbed water dehydration [21]. The final stage of mass reduction which could be detected in the temperature region of 480 – 700 °C, exhibited an endothermic peak at ~ 625 °C in DTA curve with more prominent mass reduction of ~ 23.3%. This could be recognized to the elimination of by-products from incomplete condensation of the starting materials, mostly due to the elimination of nitrate ions [22]. Thermogravimetry is a most reliable analyses used to determine the calcination temperature of dried samples derived from sol-gel technique by taking the temperature at
which the mass reduction becomes constant [23]. The TGA analysis showed that there was no mass reduction after the temperature reached 700 °C. Therefore, 700 °C temperature was chosen as calcination temperature for the dried powder.

3.2. Physical properties

The physical properties such as linear shrinkage, open porosity and relative density of sintered SiO$_2$-CaO-Na$_2$O-based ceramics were evaluated. It was observed that the linear shrinkage and relative density were found to be 2.5 ± 0.5 and 54.4 ± 0.4 respectively with the open porosity of 45.2 ± 0.7. The method of preparation and the sintering temperature plays a major role on porous nature of the ceramic materials. It has been described that the bioceramics prepared via sol-gel method showed porosity irrespective of the precursor materials. The removal of residual volatile moieties through calcination of the gel powder obtained by the sol-gel process led to the formation of pores in the final product [24, 25]. A proportional correlation between the linear shrinkage, densification and sintering temperature was well established in which the linear shrinkage and densification are increased by increasing the sintering temperature which leads to pore removal. However, the present results showed that the sol-gel synthesized SiO$_2$-CaO-Na$_2$O-based material using waste as renewable sources presented fully crystalline ceramic at a sintering temperature of 900 °C with an average porosity of 45%. The result of porosity in the present study is consistence with the earlier reported studies of sol-gel derived soda lime silica-based bioceramics [26].

3.2. X-ray diffraction (XRD)

XRD analysis was performed to study the crystallization of sodium-calcium-silicate phases in sintered ceramic specimens. The X-ray diffractogram for the ceramic specimens after sintering at 900 °C was showed in Fig. 2. It was observed that the diffractogram depicted two evident phases for the sintered samples. The major crystalline phase was
detected as combeite high (Na$_6$Ca$_3$Si$_6$O$_{18}$) (JCPDS 77-2189) [27]. It was also observed that the small amount of 16-sodium Tetracalcium Cyclo-hexasili cate (Na$_{15.6}$Ca$_{3.84}$ (Si$_{12}$O$_{36}$)) (JCPDS 75-1332) phase was detected as minor phase in the specimens. Further, crystallite size of ceramic was calculated from XRD pattern using Scherrer’s equation by taking the value of full width half maxima (FWHM) of high intense representative peaks, which gave an average crystallite size of ~ 43 nm. Fig. 3 demonstrates the X-ray diffractograms of ceramic specimens those incubated in SBF solution for different time intervals. As can be seen from Fig. 3, the crystallinity index for sodium-calcium-silicate phases reduced with an increase in soaking time of SBF solution and the new distinctive hydroxyapatite (Ca$_{10}$(PO$_4$)$_6$(OH)$_2$) peaks were observed in consistence with JCPDS no: 09-0432 [28], demonstrating that the hydroxyapatite layer mineralization on the sample after soaking in SBF solution. Furthermore, the crystallinity index for hydroxyapatite enhanced with prolonged soaking time in SBF solution.

### 3.3. FTIR spectra

Fourier transform infrared transmittance spectra of sintered SiO$_2$-CaO-Na$_2$O-based ceramics before and after soaking in SBF solution are illustrated in Fig. 4. The infrared spectra of sintered ceramic showed the most intense bands in the region of 450-1100 cm$^{-1}$ corresponded to silica network. In this region, the peaks located at ~ 623 cm$^{-1}$ and ~ 684 cm$^{-1}$ were assigned to the symmetric stretching of Si-O-Si and the peaks at ~ 1022 cm$^{-1}$ and 1075 cm$^{-1}$ corresponded to the asymmetric stretching of Si-O-Si. Further, the bands at ~ 456 cm$^{-1}$ and ~ 524 cm$^{-1}$ were characteristic to the Si-O-Si bending vibrations [29]. The band at ~ 932 cm$^{-1}$ was ascribed to non-bridging stretching oxygen bonds (NBO) of Si-O-NBO [30]. The infrared band at ~ 3434 cm$^{-1}$ was observed in sintered ceramics could be due to the stretching vibration of moisture absorption. After soaking in SBF solution, presence of new peaks at ~ 563 cm$^{-1}$ and ~ 607 cm$^{-1}$ in arrangement with the presence of peaks at ~ 1033 cm$^{-1}$ and ~
1093 cm\(^{-1}\) were evidence to the phosphate group. In particular, the dual band at \(\sim 563\) cm\(^{-1}\) and \(\sim 607\) cm\(^{-1}\) confirmed the typical feature of crystalline hydroxyl carbonated apatite (HCA) [31, 32]. Moreover, the bands resemble to the carbonate group (CO\(_3^{2-}\)) were detected at \(\sim 1418\) cm\(^{-1}\), \(\sim 1487\) cm\(^{-1}\) along with small shoulder at \(\sim 875\) cm\(^{-1}\) after soaking in SBF solution. The results specify that hydroxyl carbonate apatite (HCA) was mineralized [33]. The band at \(\sim 3434\) cm\(^{-1}\) was evidence of stretching vibration of moisture absorption, while the band at \(\sim 1642\) cm\(^{-1}\) related with absorbed H\(_2\)O bending vibration [34].

3.4. SEM-EDS analysis

The microstructural images of sintered ceramics obtained from SEM can be visualized in Fig. 5. The microstructure of sintered sample clearly showed the intergranular structure with pores. The removal of residual volatile moieties through calcination and sintering originates the pores on the surface. Microstructural observations also evidence that the formation of vermicular structure due to the together sintering of sodium calcium silicate particles. As it is known, sintering of polycrystalline ceramics ensues by diffusional transport of matter along definite paths that define the sintering mechanisms. The sintering occurs when the sodium calcium silicate particles are in contact together. Consequently, the particle sintered in contact points and result in a vermicular structure. This type of microstructural images were described by other studies in calcinated and sintered ceramics [23, 35]. Furthermore, the elemental analysis of the synthesized SiO\(_2\)-CaO-Na\(_2\)O-based bioceramic was analysed quantitatively using EDS spectra. The EDS spectra confirmed the presence of elements such as Ca, Na, Si and O.

The surface of the ceramics was exposed again to SEM analysis to identify the possible changes in their microstructure upon soaking in SBF solution as an outcome of hydroxyapatite mineralization. The corresponding images are presented in Fig. 6. The high
magnification images revealed the presence of fluffy aggregates through radially packed nanosheets on the surface of ceramics, while their quantity became higher and dense as the soaking time increased from 7 to 21 days. Furthermore, at lower magnification, the agglomerated spherical-shaped entities were evidenced. Similar micrographs were described at the surface of SBF immersed 45S5 bioglass derived glass ceramics [26, 36]. The results could represent a considerable fast and pronounced capacity of apatite mineralization of prepared SiO$_2$-CaO-Na$_2$O-based bioactive ceramics using waste as renewable sources. These statements were sustained by EDS spectra. In the elemental mapping images, the signals related with P and Ca became dominant and shielded the contributions of the elements found at the surface, emphasizing the quantitative conformation of mineralization of hydroxyapatite layer.

3.5. Variation in pH level of SBF solution

The pH changes of SBF solution with the soaking time as a result of ion exchange reactions at the solid-liquid interface are illustrated in Fig. 7. The pH of the SiO$_2$-CaO-Na$_2$O-based ceramic soaked SBF solution was shown to increase from 7.4 to 7.85, followed by initial rapid increase, with small increments with increasing soaking time. After 11 days of soaking, the pH remained relatively stable at around pH of 7.85. According to the mechanism suggested by Hench [37], the apatite mineralization was accompanying to the release of Ca$^{2+}$ and Na$^+$ ions from the ceramic sample through ion replacement method with H$^+$ and H$_3$O$^+$ ions from the SBF solution which is responsible for the increase in pH level. The ion replacement leads to the formation of SiO$_2$-rich layer on the sample surface, which provides favourable environments for mineralization of hydroxyapatite layer. Once the SiO$_2$-rich layer formed on the sample surface, the calcium and phosphate ions are migrated onto the surface from the SBF solution to grow into hydroxyapatite. In the present study, the changes in the pH obtained in the range of 7.4 - 7.85 when synthesized SiO$_2$-CaO-Na$_2$O-based ceramic was
soaked in SBF solution. The results were comparable to the pH range reported for the soda lime silica-based bioglass ceramics in earlier cases [38, 39] and it was favourable environment for bone cell culture [40].

3.6. Degradation studies

The degradation behaviour of prepared SiO$_2$-CaO-Na$_2$O-based bioactive ceramics was evaluated in SBF solution as well as in Tris-HCl buffer solution. Fig. 8 shows the degradation profile of ceramic discs after soaking in SBF and in Tris-HCl buffer solution for up to three weeks. SiO$_2$-CaO-Na$_2$O-based ceramics presented a slightly rapid degradation rate for the first 24 h than that of latter immersion period in SBF solution, and its weight loss after three weeks of soaking reached only about ~ 7.2%. This could be due to the reprecipitation of ions in the SBF solution as a result in the mineralization of hydroxyapatite. The degradation rate in Tris-HCl buffer solution was much faster compared to the degradation rate in SBF solution. The degradation rate was increased exponentially with increasing soaking time in Tris-HCl solution, and its weight loss reaching about ~ 40.3% after three weeks of soaking. Tris-HCl is a plain buffer solution that contains no ions therefore it shows minimal reprecipitation activity and maximum solubility of the material [41]. Dissolution rate is an important feature that a bioactive substance must fulfil. The material dissolution produces ions that act as enhancements to the medium stimulating osteogenesis, though the dissolution rate of a bioactive material and the kinetics of mineralization of apatite need to be in the same order for safe implantation [36]. In general, alkali-based bioactive materials possess a slightly fast dissolution rate which leads to the abrupt changes in the pH values of local physiological microenvironment [42]. The dissolution rate of these alkali-based bioceramics can be improved on ion-based modification by doping the less soluble phases like Zr, Zn, Mg, etc. On the other hand, the degradation study results of the this study exhibited that the
synthesized SiO₂-CaO-Na₂O-based ceramic using waste as renewable sources demonstrated considerable controlled dissolution rate in SBF solution.

3.7. Cytocompatibility

The in vitro biocompatibility of prepared SiO₂-CaO-Na₂O-based bioactive ceramics was analyzed using MG 63 osteoblast-like cells. Specifically, cytocompatibility was assessed by determining the percentage of cell viability 24 hours after incubation of SiO₂-CaO-Na₂O-based bioactive ceramics with osteoblast-like MG 63 cells by MTT assay. The corresponding cell viability profile is presented in Fig. 9. As can be understood from the cell viability profile, the relative percentage of cell viability slightly decreased after treating with different concentrations (31.25 μg/mL – 2000 μg/mL) of bioceramic particle, but not less than 75%. ISO 10993-5 says that the material can be considered cytotoxic when it losses 30% of its cell viability [43]. In the current study, the relative cell viability is greater than 75% even at higher concentration of bioceramics. Therefore, cytocompatibility evaluation results could demonstrate that SiO₂-CaO-Na₂O-based bioactive ceramics synthesized using waste as renewable sources have pronounced cytocompatibility potential with human osteoblast-like cells.

4. Conclusion

Soda lime silica-based (SiO₂-CaO-Na₂O) bioceramic was produced via sol-gel process, utilizing rice husk ash and eggshells as renewable sources for silica and calcium oxide, respectively. Combeite high (Na₆Ca₃Si₆O₁₈) was determined as a major crystalline phase in the sol-gel derived sintered sample at 900 °C with an average porosity of 45%. The hydroxyapatite mineralization ability of resultant ceramic was confirmed using simulated body fluid (SBF). The results of in vitro bioactivity test showed that the ceramics exhibited excellent bioactivity with mineralization of carbonated hydroxyapatite on its surface within 3
days of SBF immersion. Degradation study results showed that the soda lime silica-based ceramics thus produced has substantial controlled degradation rate. Cytocompatibility evaluation results demonstrated the pronounced cytocompatibility potential at different dosages (31.25 μg/mL – 2000 μg/mL) with human osteoblast-like MG-63 cells. Therefore, SiO₂-CaO-Na₂O-based ceramics prepared using waste as renewable sources might be prospective low cost ceramics for use in biomedical applications. The work also established believably that, the synthesis method is potential beneficial for the production of low cost soda lime silica-based bioceramics.

Acknowledgements

The authors would like to thank MHRD & Science and Engineering Research Board, India for financial assistance (Ref: Letter No. EMR/2016/006870). We also thank the Director, National Institute of Technology, Warangal for providing us with facilities to carrying out the experiments.

Funding:

Funding: This study was funded by Science and Engineering Research Board, India (grant number EMR/2016/006870)

Conflict of Interest:

Author P. Abdul Azeem declares that he has no conflict of interest. Co-author Palakurthy Srinath declares that he has no conflict of interest. Co-author K. Venugopalreddy declares that he has no conflict of interest.

Availability of data and material:

Not applicable

Code availability:
Not applicable

**Ethical approval:**

This article does not contain any studies with human participants or animals performed by any of the authors.
References

1. Hench LL (2006) The story of Bioglass®. J Mater Sci Mater Med. 17(11):967-78. doi: 10.1007/s10856-006-0432-z

2. Surmenev RA, Surmeneva MA, Ivanova AA (2014) Significance of calcium phosphate coatings for the enhancement of new bone osteogenesis - A review. Acta Biomater. 10:557-579. doi: 10.1016/j.actbio.2013.10.036

3. Madrid D (2004) Calcium phosphates as substitution of bone tissues. Progress in Solid State Chemistry. 32:1–31. doi: org/10.1016/j.progsolidstchem.2004.07.001

4. Pezzotti G (2017) Bioceramics are not bioinert. Mater Today 20:395–398. doi:org/10.1016/j.mattod.2017.06.008

5. Best SM, Porter AE, Thian ES, Huang J (2008) Bioceramics: Past, present and for the future. J Eur Ceram Soc. 28:1319-1327. doi:org/10.1016/j.jeurceramsoc.2007.12.001

6. Lin K, Wu C, Chang J (2014) Advances in synthesis of calcium phosphate crystals with controlled size and shape. Acta Biomater. 10:4071-4102. doi:org/10.1016/j.actbio.2014.06.017

7. Thamaraiselvi T, Rajeswari S (2004) Biological evaluation of bioceramic materials-a review. Trends in biomaterials & artificial organs 18. Carbon N Y 24:172

8. Vallet-Regí M, Ruiz-Hernández E (2011) Bioceramics: From bone regeneration to cancer nanomedicine. Adv. Mater. 23:5177-218. doi: 10.1002/adma.201101586.

9. Zhang Y, Santos JD (2000) Crystallization and microstructure analysis of calcium phosphate-based glass ceramics for biomedical applications. J Non Cryst Solids. 272:14-21. doi:org/10.1016/S0022-3093(00)00115-0
10. Kumar S, Vinatier P, Levasseur A, Rao KJ (2004) Investigations of structure and transport in lithium and silver borophosphate glasses. J Solid State Chem. 177: 4-5. doi:org/10.1016/j.jssc.2003.12.034

11. Jaroch DB, Clupper DC (2007) Modulation of zinc release from bioactive sol-gel derived SiO 2-CaO-ZnO glasses and ceramics. J Biomed Mater Res - Part A. 82:575-88. doi:org/10.1002/jbm.a.31180

12. Arcos D, Vallet-Regí M (2010) Sol-gel silica-based biomaterials and bone tissue regeneration. Acta Biomater. 6:2874–2888. doi:org/10.1016/j.actbio.2010.02.012

13. Lao J, Nedelec JM, Moretto P, Jallot E (2006) Micro-PIXE characterization of interactions between a sol-gel derived bioactive glass and biological fluids. Nucl Instruments Methods Phys Res Sect B Beam Interact with Mater Atoms. 245:511-518. doi:org/10.1016/j.nimb.2005.12.049

14. Ergun C, Evis Z, Webster TJ, Sahin FC (2011) Synthesis and microstructural characterization of nano-size calcium phosphates with different stoichiometry. Ceram Int. 37:971-977. doi:org/10.1016/j.ceramint.2010.11.004

15. Zheng K, Boccaccini AR (2017) Sol-gel processing of bioactive glass nanoparticles: A review. Adv. Colloid Interface Sci. 249:363-373. doi:org/10.1016/j.cis.2017.03.008

16. Abu R, Yahya R, Neon S (2016) Production of High Purity Amorphous Silica from Rice Husk. Procedia Chem 19:189–195. doi:org/10.1016/j.proche.2016.03.092

17. ASTM International (2013) Standard Test Methods for Density of Compacted or Sintered Powder Metallurgy (PM) Products Using Archimedes’ Principle. Astm B962-13 i:1–7. doi:org/10.1520/B0962-17.2
18. Kokubo T, Takadama H (2006) How useful is SBF in predicting in vivo bone bioactivity? Biomaterials 27:2907–2915. doi: 10.1016/j.biomaterials.2006.01.017

19. Kokubo T, Kushitani H, Sakka S, et al (1990) Solutions able to reproduce in vivo surface-structure changes in bioactive glass-ceramic A-W3. J Biomed Mater Res. 24:721-34. doi:org/10.1002/jbm.820240607

20. Palakurthy S, P AA, K VR (2019) In vitro evaluation of silver doped wollastonite synthesized from natural waste for biomedical applications. Ceram Int. 45:25044-51. doi:org/10.1016/j.ceramint.2019.03.169

21. Oki A, Parveen B, Hussain S, et al (2004) Preparation and in vitro bioactivity of zinc containing sol-gel-derived bioglass materials. J Biomed Mater Res - Part A. 69:216-21. doi:org/10.1002/jbm.a.20070

22. Montazerian M, Yekta BE, Marghussian VK, et al (2015) Bioactivity and cell proliferation in radiopaque gel-derived CaO-P2O5-SiO2-ZrO2 glass and glass-ceramic powders. Mater Sci Eng C. 55:436-47. doi:org/10.1016/j.msec.2015.05.065

23. Hassanzadeh-Tabrizi SA, Taheri-Nassaj E (2008) Effects of milling and calcination temperature on the compressibility and sinterability of a nanocrystalline Al2O3-Y 3Al5O12 composite powder. J Am Ceram Soc 91:3546–3551. doi:org/10.1111/j.1551-2916.2008.02727.x

24. Choudhary R, Venkatraman SK, Chatterjee A, et al (2019) Biomineralization, antibacterial activity and mechanical properties of biowaste derived diopside nanopowders. Adv Powder Technol. 30:1950-1964. doi:org/10.1016/j.apt.2019.06.014

25. Lakshmi R, Velmurugan V, Sasikumar S (2013) Preparation and Phase Evolution of Wollastonite by Sol-Gel Combustion Method Using Sucrose as the Fuel. Combust Sci
26. Du R, Chang J (2004) Preparation and characterization of bioactive sol-gel-derived \( \text{Na}_2\text{Ca}_2\text{Si}_3\text{O}_9 \). J Mater Sci Mater Med. 5:1285–1289

27. Nayak JP, Kumar S, Bera J (2010) Sol-gel synthesis of bioglass-ceramics using rice husk ash as a source for silica and its characterization. J Non Cryst Solids 356:1447–1451. doi:org/10.1016/j.jnoncrysol.2010.04.041

28. Palakurthy S, K. VGR, Samudrala RK, P. AA (2019) In vitro bioactivity and degradation behaviour of \( \beta \)-wollastonite derived from natural waste. Mater Sci Eng C 98:109-117. doi:org/10.1016/j.msec.2018.12.101

29. Fernández CA, Martínez CA, Prado MO, et al (2015) Bone Regeneration with Wharton’s Jelly-Bioceramic-Bioglass Composite. Procedia Mater Sci 9:205–212. doi:org/10.1016/j.mspro.2015.04.026

30. Li HC, Wang DG, Chen CZ (2015) Effect of zinc oxide and zirconia on structure, degradability and in vitro bioactivity of wollastonite. Ceram Int 41:10160–10169. doi:org/10.1016/j.ceramint.2015.04.117

31. Kim JH, Kim SH, Kim HK, et al (2002) Synthesis and characterization of hydroxyapatite crystals: A review study on the analytical methods. J Biomed Mater Res. 62:600-12. doi:org/10.1002/jbm.10280

32. Ohtsuki C, Kokubo T, Takatsuka K, Yamamuro T (1991) Compositional dependence of bioactivity of glasses in the system CaO-SiO2-P2O5. Its in vitro evaluation. J Ceram Soc Japan Int ed 99:2–7. doi:org/10.2109/jcersj.99.1

33. Samudrala R, Abdul Azeem P, Penugurti V, Manavathi B (2017) Cytocompatibility
studies of titania-doped calcium borosilicate bioactive glasses in-vitro. Mater Sci Eng C 77:772–779. doi:org/10.1016/j.msec.2017.03.245

34. Lin K, Zhang M, Zhai W, et al (2011) Fabrication and characterization of hydroxyapatite/wollastonite composite bioceramics with controllable properties for hard tissue repair. J Am Ceram Soc. 94:99-105. doi:org/10.1111/j.1551-2916.2010.04046.x

35. Zahedi M, Hassanzadeh-Tabrizi SA, Saffar-Teluri A (2018) Sol-gel synthesis and luminescence properties of Ba2SiO4:Sm3+ nanostructured phosphors. Ceram Int 44:10169–10174. doi:org/10.1016/j.ceramint.2018.03.006

36. Chen QZ, Thompson ID, Boccaccini AR (2006) 45S5 Bioglass s -derived glass – ceramic scaffolds for bone tissue engineering. 27:2414–2425. doi:org/10.1016/j.biomaterials.2005.11.025

37. Hench LL (1998) Bioceramics. Journal of the American Ceramic Society. 81:1705- 28. doi:10.1111/j.1151-2916.1998.tb02540.x

38. Peitl O, Dutra Zanotto E, Hench LL (2001) Highly bioactive P2O5-Na2O-CaO-SiO2glass-ceramics. J Non Cryst Solids 292:115–126. doi:org/10.1016/S0022-3093(01)00822-5

39. Abo-Mosallam HA, Salama SN, Salman SM (2009) Formulation and characterization of glass-ceramics based on Na 2Ca2Si3O9-Ca5(PO 4)3F-Mg2SiO4-system in relation to their biological activity. J Mater Sci Mater Med 20:2385–2394. doi:org/10.1007/s10856-009-3811-4

40. Galow AM, Rebl A, Koczan D, et al (2017) Increased osteoblast viability at alkaline pH in vitro provides a new perspective on bone regeneration. Biochem Biophys...
41. EN-ISO-10993-14 (2001) Biological evaluation of medical devices - Part 14: Identification and quantification of degradation products from ceramics. ISO 10993-142001

42. Fiume E, Barberi J, Vern E, Baino F (2018) Bioactive Glasses: From Parent 45S5 Composition to Scaffold-Assisted Tissue-Healing Therapies. J Funct Biomater. 9:24. doi: 10.3390/jfb9010024

43. International Organization for Standardization (2009) Biological evaluation of medical devices - Part 5: Tests for in vitro cytotoxicity. Iso 10993–5.
Figure captions

Fig. 1 Thermal analysis of dried gel powder

Fig. 2 X-ray diffractogram of ceramics after sintering at 900 °C

Fig. 3 X-ray diffractogram of ceramics after soaking in SBF solution

Fig. 4 FTIR spectra of sintered ceramic before and after soaking in SBF solution

Fig. 5 The surface microstructure and elemental analysis of sintered ceramic

Fig. 6 The surface microstructure and elemental analysis of sintered ceramic after soaking in SBF solution

Fig. 7 Change in pH of SBF solution during bioactivity study

Fig. 8 Degradation behaviour of prepared SiO$_2$-CaO-Na$_2$O-based bioactive ceramics in SBF and in Tris-HCl buffer solution

Fig. 9 Cell viability of osteoblast-like MG 63 cell when exposed to the bioceramic particles
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FTIR spectra of sintered ceramic before and after soaking in SBF solution
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The surface microstructure and elemental analysis of sintered ceramic

Figure 6

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Change in pH of SBF solution during bioactivity study
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Figure 9

Cell viability of osteoblast-like MG 63 cell when exposed to the bioceramic particles

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